**IAEA-TECDOC-1592** 

# Reference Asian Man: Ingestion and Organ Content of Trace Elements of Importance in Radiological Protection



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#### FOREWORD

National and international authorities responsible for radiological protection draw upon a wide variety of data to help them in the formulation of specific advice, codes of practice and regulations designed to protect occupational workers and members of the public from the harmful effects of ionizing radiation.

A practical concept developed more than 30 years ago for application in this field was that of 'reference man' — a formalized description of some specific physical, chemical, physiological, anatomical and biokinetic parameters of relevance in radiological protection. Earlier work on this topic had led in 1975 to the 'ICRP Reference Man' publication of the International Commission on Radiological Protection, which is still very widely used as a handbook. However, it has been recognized for many years that, because the data presented in this report are based primarily on Caucasian populations, they are not completely appropriate for application in the Asian region. It was mainly for this reason that a group of Member States within the Regional Cooperative Agreement for Research, Development and Training Related to Nuclear Science and Technology for Asia and the Pacific (RCA) decided, in 1987, with financial support from the Government of Japan, to set up a specific project on Reference Asian Man. This project, which was executed in two phases, was originally developed within the framework of the RCA project on Strengthening of Radiation Protection Infrastructures.

The first phase of the project (1988–1993), which has already been published in IAEA-TECDOC– 1005, Compilation of Anatomical, Physiological and Metabolic Characteristics for a Reference Asian Man (1998), dealt with the collection of physical, anatomical and physiological parameters for Asian populations. The second phase (1995–2000), which is the subject of this report, was conducted through a Coordinated Research Project from 1995 to 2000, supplemented by additional work lasting until 2004. Its main purpose was to obtain accurate and representative data for Asian populations on dietary intakes and organ contents of a group of elements that are important in radiological protection, mainly caesium, iodine, strontium, thorium and uranium. A few other elements were also studied, including some that are of importance in human nutrition.

Nine RCA Member States participated in this second phase of the project: Bangladesh, China, India, Indonesia, Japan, Pakistan, Philippines, Republic of Korea and Vietnam. The project was successful in producing the first-ever set of independently generated ingestion and organ content data for the elements of primary interest in these Asian countries.

It is hoped that this publication will serve as a useful handbook for radiological protection workers, particularly in the Asian region, but also elsewhere.

The IAEA wishes to thank the Government of Japan for its support of this project through the Ministry of Foreign Affairs and the National Institute of Radiological Sciences (NIRS). Special thanks are expressed to H. Kawamura of NIRS, who headed the Central Reference Laboratory that was established for the purpose of this project.

The IAEA officers responsible for this project were R.M. Parr (1995–1998) and G.V. Iyengar (1999–2004) of the Division of Human Health.

## EDITORIAL NOTE

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#### 1. SUMMARY

The accurate assessment of radiation doses to human beings from radionuclides that are encountered in nuclear fuel cycle operations calls for a knowledge of some pertinent human characteristics. A practical concept developed more than 30 years ago for application in this field was that of 'reference man' — a formalized description of some specific physical, chemical, physiological, anatomical and biokinetic parameters of relevance in radiological protection. Earlier work on this topic had led in 1975 to the 'ICRP Reference Man' publication of the International Commission on Radiological Protection, which is still very widely used as a handbook. However, it has been recognized for many years that, because the data presented in this report were based primarily on the Caucasian populations of Western Europe and North America (roughly 20% or the world population), they are not completely appropriate for application in the Asian region (more than 50% or the world population). Caucasian and Asian populations differ significantly in their physique, dietary habits and general life style. Moreover, the ICRP reference man was based on data collected more than three decades ago at a time when analytical methodologies were not sufficiently well developed for determining many of the trace elements of interest at the extremely low concentrations at which they commonly occur.

To address some of the issues raised above, a reference Asian Man Project was started by the IAEA in 1988 with financial support from the Government of Japan under the framework of the Regional Cooperative Agreement (RCA) Project on Strengthening of Radiation Protection Infrastructures. This was one of many recent initiatives taken by the IAEA to support radiation protection programmes in its Member States, such as the development of the Basic Safety Standards (BSS) and radioactive waste disposal methodologies.

The first phase of the Reference Asian Man Project (1988–1993) which was published in IAEA-TECDOC-1005, Compilation of Anatomical, Physiological and Metabolic Characteristics for a Reference Asian Man (1998) concluded with the recommendation that "additional data on specific elements related to uptake of important radionuclides in national diets and selected organs from people within the RCA region is needed for internal dose assessment in the RCA countries. Continued research, with emphasis on quality assurance, should be undertaken to obtain this information for each RCA country. This programme should be formulated using methodology and protocols already established in other IAEA nutrition studies".

This recommendation set the stage for the second phase of the project, namely the Coordinated Research Project (1995–2000) described in this report, which was followed by additional analyses and evaluations, under the supervision of a Steering Committee, lasting until 2004. The scope of the whole project covered (1) the development of appropriate analytical capabilities in the participating RCA Member States, and (2) the generation of accurate data from the Asian region on daily dietary intakes and organ contents of trace elements of importance in radiation protection and dietary intake data on nutritionally essential trace elements. Follow-up studies on applications of the data generated during the CRP to problems related to radiological protection in the Asian countries were left to the individual participants.

Nine Asian countries, namely Bangladesh, China, India, Indonesia, Japan, Pakistan, Philippines, Republic of Korea and Vietnam, which together represent more than 80% of the Asian population and around 50% of the world population, participated in the project.

Some important features were incorporated during the implementation of the CRP to ensure the generation of accurate and representative data. A Steering Committee (SC) was appointed to provide guidelines on the harmonization of sampling and sample processing protocols and also to monitor the progress of the trace element data generated. In addition, the participating country laboratory at the National Institute of Radiological Sciences (NIRS) in Japan was designated to assist the IAEA as the

Central reference Laboratory (CRL) with responsibilities for training and support, for analytical quality assurance and control (QA/QC), for backup analyses, and for data compilation and evaluation.

Highly sensitive analytical techniques were used by all participants in the CRP. The main ones were instrumental neutron activation analysis (INAA), radiochemical neutron activation analysis (RNAA), inductively-coupled plasma mass spectrometry and atomic emission spectrometry (ICP-MS and ICP-AES), and atomic absorption spectrophotometry (AAS).

About 5000 analytical determinations were made on 220 diet samples collected in the participating Asian countries, together with about 600 tissue samples and a large number of individual food specimens. Daily dietary intakes of more than 20 elements were measured. They included all of the elements of major importance in radiological protection, such as Ca, Cs, I, K, Sr, Th and U, as well as several trace elements of nutritional importance, such as Cr, Cu, Fe, Mn, Se and Zn. Some toxic elements were also studied in a few countries.

This project resulted in the first set of independently generated ingestion data for the elements of interest at normal environmental levels together with their related organ contents (this contrasts with the data reported in ICRP-23, which were compiled from the literature.) These data were also shown to be consistent with the new ICRP biokinetic models for the radionuclides <sup>137</sup>Cs, <sup>131</sup>I, <sup>90</sup>Sr, <sup>232</sup>Th and <sup>238</sup>U.

The project showed that dietary intakes of the elements of radiological importance in Asian populations are consistently lower than the intakes reported for ICRP reference man. The main feature of the results on essential elements was the large inter-country and intra-country variations in the intakes of many elements. The intakes of essential trace elements were generally lower in the Asian region than those reported for ICRP reference man, with the exception of Mn and to some extent Fe. Daily intakes of Ca and Zn in all Asian countries were consistently lower than the dietary allowances recommended by the US National Academy of Sciences. The organ content data generated in this CRP were consistently lower than the values reported by ICRP.

The research work carried out as part of the CRP has so far resulted in more than 20 scientific publications by the participants in peer-reviewed journals.

#### 2. INTRODUCTION

#### 2.1. ICRP reference man for radiological protection

Radiation monitoring of human subjects is an important component of programmes aimed at harnessing nuclear energy for peaceful purposes, such as electric power generation and nuclear medicine. In normal day-to-day operations, it is mainly the occupational worker who is exposed. But at the time of an emergency arising out of an accidental release of radioactivity (as in the case of the Chernobyl nuclear power plant accident in 1986), subjects living in the surrounding areas may also be exposed to nuclear radiation and thereby face health hazards.

The ICRP makes periodic recommendations [1, 2] concerning primary safety limits of radiation exposure to occupational workers as well as to the general public in order to ensure that the health hazards due to radiation exposure are kept within acceptable safety limits. The Commission bases its recommendations of primary limits on the harmful effects of nuclear radiation such as cancer and genetic disorders in progenies of the exposed individual. The Commission has contributed significantly in the past through its various constituent committees and the task groups set up to

provide guidelines in radiation protection. The Commission has also assisted in the development of secondary radiation protection standards such as annual limits of intake (ALI) and derived air concentrations (DAC) for important radionuclides.

The calculation of the radiation dose to occupational workers and to the general public from their exposure to radionuclides requires a knowledge of pertinent human characteristics such as physical, physiological, anatomical and biokinetic parameters. These were first provided by the ICRP in 1975 in its still widely used Report of the Task Group on Reference Man, Publication 23 [3] (henceforth, in this document, this report is referred to simply as ICRP-23). Some limitations of data included in ICRP-23 were recognized even at the time of the publication itself and even more so, later on, following the Chernobyl nuclear power plant accident in 1986.

#### 2.1.1. Limitations of ICRP Publication 23

ICRP-23 was the first serious attempt to compile pertinent human data of the kind that are needed in radiological protection. As such it was a landmark publication which is still widely used and quoted. Nevertheless, it has some limitations arising from the fact that it was compiled mostly from studies conducted in Europe and North America; it therefore mainly reflects the characteristics of the Caucasian populations of these regions which together comprise less than 20% of the world's population. A significant deficit is that it does not represent the populations of Asian countries, many of which now have significant nuclear energy programmes. Some statistics on the population sizes of the Asian countries that participated in this CRP, and the total world population, are given in Table 1.

World	6134			
China	1285			
India	1025			
Indonesia	214			
Pakistan	145			
Bangladesh	140			
Japan	127			
Philippines	77			
Vietnam	79			
Republic of Korea	47			
Total of project countries	3139			
Rest of the world other than Asia.	2413			
Source: United Nations World Urbanization Prospects [7]				

# TABLE 1: DISTRIBUTION OF POPULATIONS IN VARIOUS REGIONS AND COUNTRIES OF THE WORLD (millions)

Data reported in the last couple of decades from Asian countries such as India [4, 5] and Japan [6] have indicated beyond doubt that Asian populations have physical, anatomical and physiological characteristics which are quite different from those of the Caucasian populations represented in ICRP-23. In cases of low radiation exposure, the error in the assessment of radiation doses due to differences in human characteristics may not be of much consequence. However, when radiation exposures are high, the use of population and region specific human data becomes more important because of the greater need to measure radiation doses accurately and to adopt appropriate intervention practices.

ICRP-23 is a compilation of literature values available at the time of publication; most of the reported studies were completed prior to 1975. A serious limitation of these data is not only that they are old but also that very little analytical quality control could have been exercised at that time. An example of the non-availability of reliable data due to the lack of quality control in analytical methodologies is reflected in the estimates of the daily dietary intake of 300  $\mu$ g of Co and 7  $\mu$ g of U content in human kidney proposed for ICRP reference man [3]. These values are higher (by an order of magnitude) than the world average data recently reported by Iyengar [8] of 20  $\mu$ g Co dietary intake and 0.2  $\mu$ g U content in the kidney.

### 2.1.2. Subsequent ICRP publications and the need for additional analytical data

Realizing the shortcomings of the data in ICRP-23, a new task group was constituted in 1984. This was done to re-examine and, if found necessary, to revise the composition of reference man since, subsequent to the publication of the ICRP-23 report in 1975, substantial new data on elemental concentrations had been generated using advanced and reliable analytical techniques. Unfortunately, the work of this task group was terminated before it had completed its evaluations. However, in 1995, revised basic anatomical and physiological data for the skeleton were published in ICRP-70 [9]. The skeleton weights were revised from 10 kg and 7.5 kg for the male and female respectively (ICRP-23) to 10.5 kg and 7.8 kg (ICRP-70). Similarly, the Ca content of the human body was revised from 1.0 kg and 0.8 kg to 1.18 kg and 0.86 kg, respectively, for the two genders.

For other elements, an evaluation of the data collected during the task group duration and shortly thereafter resulted in a review published by Iyengar in 1998 [8]. However, even in this report, there were still some gaps requiring accurate data, specifically for elements of importance in radiological protection. The present CRP is therefore, in some respects, a timely response to these needs.

Shortly after completion of the project described in this report, ICRP issued a new set of recommendations in 2003 on basic anatomical and physiological data for use in radiological protection. This new document, Publication 89, is discussed in more detail in Section 11 (Addendum).

As discussed in this Addendum, there is very little commonality between ICRP Publication 89 and the present IAEA project. The ICRP publication focuses mainly on major elements such as H, C, N, O, Na, P, S, Cl and K in various soft tissues, Ca in the skeleton and kidney, Fe in the blood and I in the thyroid. Data for ingestion intake is not discussed at all. Only iodine in the thyroid is common to both reports. For further details, the reader is referred to Sections 8.2 and 11 (Addendum) of this report.

#### 2.2. Phase I of the reference Asian Man study

Realizing the need for the population and region (geographical) specific data on human characteristics from Asia for reliable internal radiation dose estimation, the National Institute of Radiological Sciences (NIRS) in Japan proposed to the IAEA a project on Reference Asian Man (RAM) for strengthening radiation protection infrastructure in the Asian region at the Project Formulation Meeting in 1987 held in Tokyo, Japan. The paucity of the data on human characteristics in the Asian region became the driving force and phase-I of the RAM project, namely 'Compilation of anatomical,

physiological and metabolic characteristics for a Reference Asian Man', was initiated and the related research work was carried out from 1988 to 1993. During this period, a comprehensive database was created on physical measurements of both male and female subjects along with the masses of their internal organs. Some data on daily dietary intakes of some elements were also compiled but were preliminary in nature and insufficient both in quality and quantity.

The final report on the first phase of the project [10] made a specific recommendation which stated: "Additional data on specific elements related to uptake of important radionuclides from national diets and their content in selected organs from people within the RCA region are needed for internal dose assessment in the RCA countries. Continued research, with emphasis on quality assurance, should be undertaken to obtain this information for each RCA country". This programme, as stated in the report, should be formulated using methodology and protocols already established in other nutrition-related studies conducted by the IAEA.

The collection of accurate and representative data on daily intakes and organ content of various trace elements of importance in radiological protection is important for estimating realistic biokinetic parameters of these elements and their related radionuclides. In turn, these are the contributing factors in making accurate internal dose assessments and in formulating secondary radiation protection standards such as annual limits on their intakes (ALI). The ingestion and organ content of trace elements such as Cs, I, Sr, Th and U, which are related to radionuclides that are encountered in the middle and front-end of the nuclear fuel cycle, were proposed to be studied in the Asian region. These data can be used for the estimation of biokinetic parameters such as the gastrointestinal absorption factor ( $f_1$ ), the organ uptake factor ( $f_2$ ) and the biological half-life ( $T_{b1/2}$ ) of the elements and their corresponding radionuclides.

#### 2.3. Initiation of the Reference Asian Man Phase II study

The decision to start the second phase of the RAM project dealing with the 'Ingestion and Organ Content of Trace Elements of Importance in Radiological Protection' emerged from a consultants' meeting that took place during 1994 at NIRS, Chiba, Japan. This was followed by a Project Formulation Meeting in 1995 in Japan, where the actual course of the CRP was decided and the protocol for sampling was discussed and partly formulated [11].

It was considered important to harmonize both the collection and processing of samples, followed by strict quality control (both internal and external) during the analysis. The decision to include Cs, I, Sr, Th and U as well as Ca and K (because of their chemical similarity to Sr and Cs, respectively) was based primarily on their relevance to the programme of studying the biokinetic behaviour of selected radionuclides encountered in nuclear fuel cycle operations. It was agreed that both tissue and dietary samples would be studied. It was also agreed upon that the daily intakes of essential trace elements of importance in human nutrition should be obtained using the same diet samples collected under well defined conditions. In all, nine countries, Bangladesh, China, India, Indonesia, Japan, Pakistan, Philippines, Republic of Korea and Vietnam, participated in the project. These countries represent about 80% of the Asian population and more than 50% of the world population as seen in Table 1.

#### 2.3.1. The need for biokinetic data of important radionuclides

The CRP had the prime objective of obtaining dietary intake and organ content data which could be used for biokinetic modelling of five important radionuclides, namely <sup>137</sup>Cs, <sup>131</sup>I, <sup>90</sup>Sr, <sup>232</sup>Th and <sup>238</sup>U, encountered at the front and middle-end of the nuclear fuel cycle. <sup>232</sup>Th is used in fast breeder reactors for the generation of fissile <sup>233</sup>U by the <sup>232</sup>Th(n, $\gamma$ )<sup>233</sup>U reaction. <sup>238</sup>U is the major component of both natural uranium and enriched uranium fuel for nuclear reactors. The other three radionuclides, <sup>137</sup>Cs, <sup>131</sup>I and <sup>90</sup>Sr, are fission products formed during fuel burn-up. The behaviour of these radionuclides in the human system can be understood through the study of the behaviour of the trace elements Cs, I, Sr, Th and U, which are present naturally in daily diets as well as in human body organs. Accurate data on

some biokinetic parameters such as the  $T_{b1/2}$ ,  $f_1$  and  $f_2$  factors for the radionuclides, are required for obtaining their dose coefficients (internal radiation dose per unit intake of the activity) employed in the calculation of the internal dose. A significant alteration in the daily intake of an element or a radionuclide may affect its dynamic equilibrium in the body and thus affect its biological half-life. This phenomenon was amply demonstrated in the data obtained in the first phase of the project. Dang et al. [12] predicted a shorter biological half-life for <sup>3</sup>H in the body of adult Indians in comparison with the Caucasians represented by ICRP reference man [13]. This work was supported by the observations of a short half-life in Indian radiation workers by Rudran et al. [14]. A number of workers have used data on daily dietary intakes, organ contents and daily urinary excretion to estimate some of the important biokinetic parameters [15–18].

Some of the above stated population specific biokinetic parameters for <sup>137</sup>Cs, <sup>131</sup>I, <sup>90</sup>Sr, <sup>232</sup>Th and <sup>238</sup>U could therefore be obtained through the study of daily ingestion, along with the organ contents of their stable counterparts Cs, I, Sr, Th and U, respectively. This is the case with radionuclides <sup>137</sup>Cs and <sup>131</sup>I, whose models were not revised and continue to be same as given in ICRP-30. However, in the case of the radionuclides <sup>232</sup>Th, <sup>90</sup>Sr and <sup>238</sup>U, simple models using single exponentials have been replaced by the more complex re-circulation models described in recent ICRP Publications [19–21]. Data on daily intakes, along with the new models, can be used to derive the organ burdens of these elements. An assessment of the agreement between the calculated and measured organ contents can demonstrate the applicability of the new ICRP models for reliable dose estimation.

The daily dietary intake also influences and determines the gastrointestinal absorption factor  $(f_1)$  of a radionuclide in the human body. The altered intake of Ca could influence both the absorption of calcium and also of the fission product <sup>90</sup>Sr. An extremely low intake of Ca has been reported to increase the gut uptake of Sr substantially [19].

#### 2.3.2. Daily dietary intake of essential trace elements for populations in the Asian Region

In view of the effort put into ensuring the generation of quality data from the Asian region, it was decided to take advantage of the possibility of using the same diet samples, collected and processed under harmonized conditions, for estimating daily intakes of some essential trace elements such as Cr, Cu, Fe, Mn, Se and Zn.

Another reason for studying intakes of essential elements, within the context of this CRP, was the reported role of essential trace elements such as Cu, Fe, Mn, Se and Zn in positively influencing the radiation dose-damage relation [22–24]. These trace elements have been shown, through the study of the biochemical radiation damage markers, to reduce the harmful effects of radiation in experimental animals. Non-toxic doses of these elements are effective in increasing the survival, and also in repairing radiation damage, when administered before irradiation. Data on intakes of essential elements could therefore be useful in future in studying the relation between the status of essential elements and their role in radioprotection of exposed individuals.

#### 2.3.3. Identification of food materials that concentrate different radionuclides

The analysis of food ingredients such as cereals, pulses, vegetables, milk, fruits, flesh foods, etc. was carried out only in India and China, and to some extent in the Republic of Korea, to identify the food ingredients that contribute most to the daily dietary intakes of the elements of interest. These data are important in radiological protection. In the case of an accidental release of radioactivity, the ingestion could well become a source of radiation exposure. In such circumstances, there may be a need to identify food components which are potential radioactivity carriers. This information could prove useful in taking relevant decisions to control the radiation exposures to humans through food materials and food chains.

#### 2.3.4. Study on organ contents of elements important in radiological protection

In addition to the intakes of trace elements, tissue samples obtained from important human organs such as lung, liver, kidney, skeleton, skeletal muscle and thyroid were also analysed for one or more of the five elements of importance in radiological protection. The choice of the tissue and the related element depended upon the known reported tendency of the organ/tissue to accumulate one or more of these five elements or their radioactive counterparts. For example, more than 90% of the iodine in the body is present in the thyroid, whereas Th tends to accumulate in bone and U in the kidney [13]. Cs is uniformly distributed in the entire soft tissue compartment, so its maximum content is in the compartment of skeletal muscle. Sr is chemically similar to Ca, so stable Sr and also fission product <sup>90</sup>Sr are deposited in the skeleton. Therefore, iodine was needed to be determined in thyroid, Cs in skeletal muscle, Th in the skeleton, lung and liver, U mainly in the skeleton, lung, kidney and liver and finally Sr was measured only in the skeleton. (Some participants however produced additional data on these trace elements in as many organs as they could determine.)

#### 2.4. Scope of the CRP

The scope of the CRP was designed to take account of the observations mentioned above, with emphasis on the following tasks:

- Establishment of protocols for collection of representative diet and tissue samples from each Asian country participating in the CRP, and processing these samples.
- Development of analytical methods for the determination of selected trace elements in diet and tissue samples and their validation through internal and external quality control.
- Analysis of (i) diet samples, (ii) food ingredients which contribute to the preparation of the diet of a country, and (iii) human tissues.
- Establishment of baseline data on (i) dietary intakes and organ contents of trace elements of importance in radiation protection, and (ii) dietary intakes of trace elements of importance in nutrition.
- Comparison of data obtained for different countries with literature values.
- Examination of the options for applying these data in radiological protection and in nutrition.

#### 3. FORMULATION OF THE CRP

The project formulation meeting was held in early 1995 at Hitachinaka City in Japan, where the actual programme of studying Cs, I, Sr, Th and U, and also the elements Ca and K, was decided and the required protocols for sampling and sample handling steps were discussed and partly formulated.

#### 3.1. Objectives of the CRP

The main objective defined for the CRP was to obtain representative data on some stable elements that are of particular importance in radiological protection models, namely the dietary intakes of Ca, Cs, I, K, Sr, Th and U and the organ contents of Cs, I, Sr, Th and U. The idea was to establish representative values for these elements in Asian populations. As a secondary objective it was also decided to collect data on the dietary intakes of nutritionally essential trace elements such as Cu, Co, Fe, Mn, Zn, etc., thereby taking advantage of the expertise on sampling, sample collection and analysis developed by the CRP participants during the course of the project.

#### 3.2. Expected output from the CRP

The research work carried out by different participating country laboratories was expected to give the following outputs:

- Improved knowledge in the Asian region of the ingestion and body content of some trace elements that are of high priority for radiological protection (by generation of new reliable data).
- Strengthened research capabilities related to sampling, analysis and data interpretation in the participating country laboratories or institutes.
- Making available new analytical Reference Materials (RMs) for the elements of interest in cases where no existing RMs were available.
- Developing applications of the data on intakes and organ contents in radiological protection.

#### 3.3. General framework of the CRP

It was agreed that as many Asian Member States as possible should be encouraged to participate in the project (subject to their membership of RCA and to screening and review of the research proposals at the IAEA). The analysis of Ca, Cs, I, K, Sr, Th and U in diets was mandatory. Optionally the participants were encouraged to undertake studies on the organ contents of Cs, I, Sr, Th and U and also to generate data on the nutritionally important trace elements such as Co, Cu, Fe, Mn, Zn, etc. For those countries which were not in a position to collect data on autopsy samples, the generation of data on the second priority elements assumed greater importance.

#### 3.3.1. Duration of the CRP and study group

The CRP was proposed to be operational from for a duration of 5 years, subject to satisfactory progress at the end of the third year.

It was agreed upon that only one study group representing the general population in each participating country was to be included; samples were to be chosen as carefully as possible with a view to making them fully representative. The age range of the subjects was decided to be 20 to 50 years.

#### 3.3.2. Samples studied

Samples chosen for the project included diets, for the ingestion part of the study, and tissue samples for determining the organ contents. The number of samples was to be sufficient to meet the objective of representative sampling. It was provisionally decided to include at least 20–30 samples of each type.

For the ingestion study, the following samples were considered: (i) representative samples of the total diet, (ii) additional duplicate diet samples to capture the variability in the intake for a country's population, and (iii) food ingredients or components (food groups) which constituted the diet.

Organ samples comprised autopsy specimens of liver (for Th and U), skeletal muscle and some other soft tissues (for Cs), rib or iliac crest and/or long bone (for Sr, Th and U) and kidney (for Th and U). The collection of the data for any additional elements in these tissues was left to the discretion of the

individual participating laboratory. It was also decided to include lung (non-systemic organ) as an optional organ for studying the contents of Th and U.

#### 3.3.3. Elements studied

These were divided into two groups. The first group (priority 1) included, Cs, I, Sr, Th and U, and also the elements Ca and K. The first five elements have radioactive counterparts (radionuclides), which are encountered in different operations of the nuclear fuel cycle. Ca and K are chemically similar to Sr and Cs respectively and therefore to the radionuclides  $^{90}$ Sr and  $^{137}$ Cs.

The second group (priority 2) included elements of nutritional importance. Depending on the availability of resources, participants carried out studies on essential minor elements such as Mg, Na, P and essential trace elements such as Cu, Fe, Mn, Se, Zn, etc. Data on elements analysed and reported by four or more countries were considered of sufficient information value to be included in this document.

#### **3.4.** Sampling strategies

The participants collaborated with national experts on nutrition and bio-statistics within individual countries to ensure that samples collected by them were representative of the population of their country. They carried out the sampling of subjects in such a way that, along with the representative intakes, it was also possible to capture the variability in the intakes of trace elements due to differences in food supply, customs, and geo-chemical changes that contributed to the differences in the intake pattern within an individual country. Details of the sampling methods used, and the analytical methods employed for various elements in different participating countries are given in Table 2. Details are discussed in Section 4.

Diet samples were prepared, based on the market basket survey or the duplicate diet method. Each participant developed written protocols, which were based on the information disseminated by the IAEA on this subject. The purpose behind this step was to ensure harmonization of the sampling practices in the participating countries. A record of the samples collected was maintained in the format shown in Table 3.

Tissue samples were collected only from subjects who had suffered a sudden accidental death (e.g. a road accident) but were otherwise healthy at the time of death. It was ensured that an autopsy was performed on these subjects within 24 hours of death. The harmonized procedures developed by the IAEA were adopted by all the countries participating in the CRP. A record of every tissue sample collected was maintained by the country laboratory. The specimen sample record sheet for autopsy specimens is shown in Table 4.

#### 3.5. Sample storage

Long term storage of biological samples was one of the requirements in order to bridge the time gap between the preparation of samples and their analysis and also for the replicate or confirmatory analysis at a later time. Short term storage of fresh samples prior to drying was done at  $-20^{\circ}$ C. Long term storage was done after homogenization and freeze drying.

Country	Sampling method	Ca	Cs	Ι	К	Sr	Th	U	Other Elements
Bangladesh	TD	AAS	ICP-MS*	-	AAS	ICP-MS*	ICP-MS*	ICP-MS*	AAS
China	TD	ICP-AES	ICP-MS <sup>†</sup>	ENAA	AAS	$ICP-AES^{\dagger}$	$\text{ICP-MS}^{\dagger}$	$ICP-MS^{\dagger}$	ICP-AES
India	TD	INAA	INAA	ENAA	INAA	RNAA	RNAA	RNAA	INAA
Indonesia	DD/TD	AAS	INAA	-	AAS	AAS	-	-	AAS
Japan	DD	ICP-AES/ INAA	ICP-MS	ICP-MS/ ENAA	ICP-AES/ INAA	ICP-AES	ICP-MS	ICP-MS	ICP-AES
Republic of Korea	TD	INAA/ICP- AES	INAA	INAA/ ENAA	INAA	ICP-AES/ RNAA	RNAA/ ICP-MS	RNAA/ ICP-MS	INAA
Pakistan	TD	AAS/ICP- AES	INAA	ENAA/ INAA	AAS/ICP- AES	ICP-AES	INAA	ICP-MS*	INAA/ AAS
Philippines	TD/DD	ICP-AES <sup>‡</sup>	ICP-MS <sup>‡</sup>	ENAA/ INAA	ICP-AES <sup>‡</sup>	ICP-AES <sup>‡</sup>	ICP-MS <sup>‡</sup>	ICP-MS <sup>‡</sup>	ICP-AES
Vietnam	TD/DD	INAA/ ICP- AES <sup>‡</sup>	INAA/ ICP-MS <sup>‡</sup>	RNAA	INAA/ ICP-AES <sup>‡</sup>	RNAA/ ICP-MS <sup>‡</sup>	RNAA/ ICP-MS <sup>‡</sup>	RNAA/ ICP-MS <sup>‡</sup>	INAA/ RNAA/ ICP-AES

# TABLE 2. SAMPLING AND ANALYTICAL METHODS USED BY PARTICIPATING COUNTRIES IN THE INGESTION STUDIES

AAS — Atomic absorption spectrometry; DD — Duplicate diet study; ENAA — Epithermal neutron activation analysis; ICP-AES — Inductively coupled plasma-atomic emissión spectrometry; ICP-MS — Inductively coupled plasma mass spectrometry; INAA — Instrumental neutron activation analysis; RANA — Radiochemical neutron activation analysis; TD — Total diet study based on market basket survey. \* By the CRL. † Diet samples in parallel by the CRL. ‡ At the CRL.

#### 3.6. Appointment of a Central Reference Laboratory

The suitability of various techniques for the determination of the elements of primary importance or first priority is shown in Tables 5(a)-5(e). It was observed that for Ca, Cu, Fe, K, Mg, Mn, Na, Zn, etc., common techniques such as AAS, which were available in most of the country laboratories, were adequate to perform the analysis. For determining Cs, I, Sr, Th, and U, in both diet and tissue samples, highly sensitive and reliable analytical techniques such as INAA, RNAA, ICP-MS and ICP-AES were developed.

#### 3.7. Appointment of a Central Reference Laboratory

During the project formulation stage of the CRP, the appointment of a Central reference Laboratory (CRL) was considered to be a crucial strategy to ensure the generation of reliable analytical data. The laboratory of the CRP participant at the National Institute of Radiological Sciences (NIRS), Japan, undertook this role. The names of individual scientists at the CRL are listed in Annex 1. In addition to providing technical advice and training, the CRL was assigned responsibilities for both internal and external quality control, together with the central compilation and evaluation of data submitted by all the CRP participants. The concept and the exact role of the CRL are described in more detail later in Section 6.

#### TABLE 3. SAMPLE RECORD FOR THE DIET SAMPLES

A: Sample Code:

**B**: General Information:

 Name of the country Geographic region within the country Specific Location (name), if known Population classification: 
 Urban 
 Rural 
 Representative Socioeconomic status: 
 Low 
 Middle 
 High Sample collection by (initials):
 Date of sample collection:

C. Type of diet collected: 
□ Market basket 
□ Duplicate diet

D. Components of the diet sample (based on FAO grouping) in grams per day:

□ Cereals □ Beans/Nuts □ Yam products □ Meat products □ Egg products □ Aquatic food □ Milk products □ Vegetables □ Fruits □ Sugar □ Drinks and water □ Alcohol in beverages

E. Preparation procedures :

Blender supplied by CRL used:	□ yes	□ no
Separate water sample collected:	□ yes	$\square$ no

- F. Diet weights:

G. Rough estimate of the caloric content of the diet, if known (Kcal/day) :

Additional information (use separate page if needed)

### TABLE 4. SPECIMEN SAMPLE RECORD SHEET FOR AUTOPSY SPECIMENS

	e.g. PHI-LV-07 (see Table 6)
SAMPLE COLLECTION	٨:
Person responsit	ble:
Where collected	:
Date:	
Time after death	hours
SUBJECT:	
Sex	
Age	
Weight (kg)	
Height (cm)	
Ethnic group	ot's name if known comments on apparent health status, etc.
Kemarks (Subjec	et s'hanne, it known, comments on apparent nearth status, etc.
SAMPLE HANDLING:	
SAMPLE HANDLING: Person responsit	ble
SAMPLE HANDLING: Person responsit Sample size (fre	ble sh) g
SAMPLE HANDLING: Person responsit Sample size (fre Sample size (dry	ble sh) g

Aliquot No.	Date	Weight (g)	Transferred to	Purpose
1				
1				
etc.				

# TABLE 5. APPLICABILITY OF THE MAIN ANALYTICAL TECHNIQUES FOR THE ELEMENTS AND SPECIMENS OF INTEREST

TABLE 5(a). APPLICABILITY	OF INAA (SAMPLE SIZE	250–500 mg DRY MA	(TER)
		U	

Sampla	Element							
Sample -	Cs	Ι	Sr	Th	U			
Bone			-	-	-			
Diet	+++	+(1)	-	+	-			
Kidney				-	-			
Liver				++				
Lung				+				
Muscle	++							
Thyroid		+++(1)						
(1) Epithermal neutron activation analysis								

### TABLE 5(b). APPLICABILITY OF RNAA (SAMPLE SIZE 250 – 500 mg DRY MATTER)

Somulo			Element		
Sample	Cs	Ι	Sr	Th	U
Bone			+++	+++	++
Diet	+++	+++	+++	+++	+++
Kidney				+++	+++
Liver				++	
Lung	+++			+++	+++
Muscle	+++				
Thyroid		+++			

- not possible + possible but with some difficulty ++ good method +++ reference method

Comple	Element								
Sample	Cs	Ι	Sr	Th	U				
Bone			+	+++	++				
Diet	+++	-	+	+++	+++				
Kidney				+++	+++				
Liver				+++					
Lung	+++			+++	+++				
Muscle	+++								
Thyroid		-							

## TABLE 5(c). APPLICABILITY OF ICP-MS (SAMPLE SIZE: 0.1–1 g DRY MATTER)

## TABLE 5(d). APPLICABILITY OF ICP-AES (SAMPLE SIZE 0.1–1. g DRY MATTER)

Sample -	Element								
	Cs	Ι	Sr	Th	U				
Bone			+++	-	-				
Diet	-	-	+++	-	-				
Kidney				-	-				
Liver				-					
Lung	-			-	-				
Muscle	-								
Thyroid		-							

- not possible + possible but with some difficulty ++ good method +++ reference method

Sample	Element								
	Cs	Ι	Sr	Th	U				
Bone			+ (*)	-	-				
Diet	++	-	++ (*)	-	-				
Kidney				-	-				
Liver				-					
Lung	+			-	-				
Muscle	+								
Thyroid		-							

(\*) Graphite furnace AAS

- not possible + possible but with some difficulty ++ good method +++ reference method

#### 3.8. Appointment of a Steering Committee

The idea of appointing a steering committee (SC) to provide general scientific guidance of the CRP was recognized and put into practice. Membership of the SC is listed in Annex 1.

The first task the SC performed was to obtain up-to-date information on the availability of suitable certified reference materials (CRMs) for the trace elements of interest and to design a scheme for their use in the present CRP. The SC also advised the IAEA and host institutes on various matters connected with quality control (QC) and data evaluation. It also assisted the IAEA with the drafting and review of this report.

The SC conducted most of its work by correspondence. However, the committee also met several times during the course of the project.

#### 3.9. Supplementary programmes

As per the guidelines issued by the IAEA, the participants developed supplementary programmes within their projects without diverting significant resources from the core programme. One such area of potential interest was the generation of input data for biokinetic models; another was to use this data to test the applicability of the new ICRP models of radiological elements at the environmental level of concentrations in diet and tissues.

#### 3.10. Data reporting and evaluation

To avoid difficulties at the time of evaluation and comparison of the data on diets and tissue materials, it was considered important that data were collected and reported in a uniform format by all the country laboratories. Data sheets in the format shown in Table 6 were used for this purpose. The final responsibility of evaluating the data collected in various Asian countries was assigned to the CRL.

#### 3.11. Use of reference materials (RMs)

For the elements Ca and K, existing RMs were found to be adequate for the concentration ranges anticipated for them. For Cs and Sr, the diet RM matrices were lacking in certified results. For I, a number of Standard reference Materials (SRMs) from the US National Institute of Standards and Technology (NIST) were used.

Th and U presented sufficient challenges for participating laboratories. Although several diverse environmental and botanical matrices were available, very few had certified values. Moreover, foods and total diet type of matrices lacked certified results. For some of these, improved reference values had to be obtained through expansion of the scope of CRP. One such example was the re-evaluation of NIST SRM 1548 total diet (see Section 6.1).

The analysis of essential elements Cu, Fe, Mg, Mn, Na, Zn, etc., presented little difficulty since they were present in diets as well as in tissues in abundant quantities and there was no difficulty in obtaining useful CRMs. NIST RMs such as bovine liver and some of the total diet RMs, which already had certified values, were effectively used for internal quality control.

#### 4. SAMPLING AND SAMPLE PREPARATION

#### 4.1. Sample collection

The success of any study related to the analysis of biological samples depends greatly on precautions taken during sample collection, storage, processing, and subsequent analysis for various constituents. Sampling and sample preparation are as important in obtaining reliable results as is the analytical methodology itself. For a long time, and in most of the programmes in the past, sufficient emphasis was placed upon accurate analysis of samples by taking care of the required quality control steps and by the use of highly sensitive analytical techniques. However decisions related to sampling and post sampling steps were often not sufficient to meet the objectives. In the present CRP, as much attention was paid to the pre-analysis steps as to the analytical part. Proper sampling protocols for harmonization of the sampling strategies were adopted in various participating countries for representative sampling of both diet and human tissues samples. Sufficient care was also taken to ensure that samples were free from extraneous contamination.

Methods employed for collecting samples were expected to meet the following objectives: (i) food samples to be truly representative of the dietary consumption of the main population of a country, (ii) sampling methodology to ensure the exclusion of external contamination, (iii) tissue samples to represent the elemental content of the whole organ and not the content of the small tissue sample itself. Specific guidelines on sampling as described below were made available to country laboratories.

#### 4.1.1. Representative sampling of populations

It was agreed upon by all participants at the very beginning of the project that sample collection would be done in a manner that ensured representative sampling. The CRP participants were therefore required to collaborate with their own national experts in nutrition to devise a plausible sampling strategy that took into account (keeping the practical constraints in focus) all the relevant ethnic, socioeconomic and geographic variables. Such representative sampling was considered important for all aspects of the CRP and particularly for the dietary intake studies.

TABLE 6: SPECIMEN DATA SHEET FOR CENTRAL REPORTING OF THE ANALYTICAL DATA (The data will be reported and recorded in the form of a spreadsheet database (e.g. Excel) with the following structure)

Sample code	Analyst code	Method	Batch	Date (YYMM)	Analyte	Unit	Value	М	RSD%
PHI-Lv-07	IA4	ICPMS	4	9704	Th	ng/kg	2.6		15
PHI-Lv-07	IA4	ICPMS	4	9704	U	ng/kg	1.5	<	
IND-Dt-17	Id1	RNAA	5	9705	Th	ng/kg	3.4	0	
etc.									

The column headings are as follows:

Sample code: This is a 3-component sample code with information on the country of origin of the samples (e.g. the Philippines in the above example) followed by the tissue identifier (Lv = liver in the above example) followed by a running number for the sample (1.99).

	Country codes		Sample codes
BGD	Bangladesh	BI	Bone (iliac crest)
CPR	China	BL	Bone (long bone)
IND	India	BR	Bone (rib)
INS	Indonesia	BS	Bone (skull)
JPN	Japan	BV	Bone (vertebra)
ROK	Republic of Korea	Dt	Diet
MAL	Malaysia	Kd	Kidney
PAK	Pakistan	Lg	Lung
PHI	Philippines	Lv	Liver
VIE	Vietnam	Ms	Muscle
		Ту	Thyroid

Analyst code: A three-letter country code followed by a running number for the analyst. In addition to the codes listed above, IA will be used to indicate the IAEA's Laboratory.

Column M contains a data Modifier indicating how the value is to be interpreted (< means that it is a "less than value; () means that the value is to be interpreted as being in brackets, i.e. that it is between the limit of detection and the limit of quantitation.

Subjects included in the study were chosen so as to capture the full range of variability associated with the intakes by different ethnic and socio-economic groups, as well as with different sources of food supply (such as those depending on the geo-chemical environment in which the food plants were grown). Other sources of variability associated with individual daily variations in intake, or with seasonal factors were generally not of interest in the study. When using the duplicate diet method, a large number of samples were required to be collected. Most of the countries therefore resorted to collection of diet samples based upon market basket survey where a relatively smaller number of samples was sufficient to obtain representative data.

It is important to understand that the populations of Asian countries are heterogeneous in their food habits and that many of the diets consumed there are primarily of plant origin (and mostly also locally grown). Therefore, either it was necessary to select not just one but several different target population groups in each country or to use country-wide data on dietary intake generated by national institutes of nutrition. Representative data in individual countries which reflected the average food consumption by the entire population was generally obtained by giving due weight to per capita income, nutritional status, per capita calorie consumption and also height and weight data for the representative population.

Pathologists and medical staff in charge of autopsy services in hospitals assisted the investigators in the collection of tissue samples. Whereas the pathologists certified that the tissue samples had no obvious abnormalities, the other medical staff ensured from hospital records that the victims had died of sudden accidental death, that they were otherwise healthy at the time of death, and that the autopsy was performed on them within 24 hours subsequent to it.

#### 4.1.2. Sampling of food and diet

Three approaches were commonly adopted when collecting food or diet samples:

- a) The first one belonged to the category of direct methods, wherein either precise weighing of the actual food consumed during the full day (24 hours) is carried out or duplicate portion of diet is collected or the total diet studies based on market basket survey are carried out.
- b) The second method involved the preparation of the balance sheet and per capita consumption or the 24-hour and 7-day recall of diets consumed. This method is often referred to as the indirect method.
- c) The last approach deals with the estimation of the food consumption based on the study of biological markers using human specimens such as urine, faeces, whole blood, etc.

Different approaches are susceptible to different kinds of errors. Whereas the first and second method may lead to underestimation or overestimation of intakes, the method dealing with the application of the biological markers could easily get affected by variable metabolic characteristics of humans. Abdulla et al. carried out a critical evaluation of various dietary sampling techniques employed for trace element studies [25].

Total diet samples studied in the CRP were prepared as 'total diet' samples (as the first preference) or as duplicate diet (second preference). However, a given participant country exercised its choice of the method depending upon its experience and the homogeneity of its population. The use of the methodologies based on food consumption tables was not adopted since reliable tables for the elements which were included in the CRP did not exist. Those countries that adopted the approach of the total diet study also collected a few duplicate diets with a view to capturing the variability in the food consumption, which in turn affected the intake of trace elements. Diet collection methods employed in the different participating countries are already described in Table 2 along with analytical techniques used for the analysis of the individual elements.

A harmonized protocol was also proposed during the first Research Co-ordination Meeting (RCM) held in Taiyuan, China, in 1996. Sample forms which the individual participants were required to complete are reproduced as Tables 3, 4 and 6.

#### 4.2. Special problems in sampling

#### 4.2.1. Need for washing the samples

The kind of precautions required during the preparation and collection of diet samples are based upon the objective of the study itself. In the study on dietary intake of essential trace elements such as Cu, Fe, Zn among others, under varied working conditions, extraneous contamination of the food material with same elements may not significantly influence the end result because of the net high intake of these elements from the food compartment.

In the present study, which also involved the application of the data intake on trace elements of importance in radiological protection to estimate their absorption through the gastrointestinal tract, contamination could adversely affect the conclusion of the study. For example, Th and U present in the food (organically bound to food) are likely to have a higher absorption through the gut than Th and U present in dust as their oxides — the source from which their absorption could be significantly lower. Dang et al. [26] have discussed the undesirable effect of dust contamination in the measurement of Th in dietary samples. Hence, any Th or U present in diet and having its origin in dust instead of food may alter the end result as well as the conclusion.

It was therefore important that food samples were very carefully washed to remove dust to minimize the entry of Th and U as extraneous contaminants. In one of the participating laboratories (India), a careful study on the concentration of six elements namely, Cr, Cs, Fe, Sr, Th and Zn in washed and unwashed food samples of cereals and legumes was conducted. It showed that, with the exception of Zn, there were statistically significant differences in the concentrations of the other five elements in washed and unwashed samples [27]. As seen in Table 7, concentrations were significantly higher in unwashed samples.

Rice			W	heat	Pulses	
Element	Unwashed	Washed	Unwashed	Washed	Unwashed	Washed
Fe (µg/g)	$5.6 \pm 0.6$	$2.5 \pm 0.3$	47.3 ± 2.3	38.6 ± 2.9	37.6 ± 2.1	33.1 ± 1.2
Zn (µg/g)	$11.4\pm0.8$	$10.8 \pm 1.1$	$21.7\pm1.9$	22.3 ± 2.1	$25.4\pm2.0$	$25.2 \pm 1.6$
Th (ng/g)	$0.9\pm0.06$	$0.6 \pm 0.03$	$3.9\pm0.3$	$1.6\pm0.2$	$1.7\pm0.2$	$0.9 \pm 0.2$
Cs (ng/g)	4.1 ± 0.3	$2.6 \pm 0.3$	$4.8\pm0.2$	$3.7\pm0.3$	44.5 ± 1.8	$37.0 \pm 2.4$
Sr (µg/g)	$0.21\pm0.05$	$0.13\pm0.01$	$5.1\pm0.7$	$3.9\pm0.5$	$3.6 \pm 0.4$	$2.5\pm0.3$
Cr (ng/g)	19.0 ± 1.8	7.9 ± <u>1</u> .0	37.1 ± 2.0	$26.5\pm1.5$	-	-

# TABLE 7. A COMPARISON OF THE CONCENTRATIONS OF TRACE ELEMENTS IN SOME UNWASHED AND WASHED RAW FOODS

#### 4.2.2. Contribution of drinking water to the daily trace element intake

Drinking water, which is consumed in its natural form or in the preparation of food and beverages, generally does not contribute significantly (<10%) to the total intake of trace elements in the diet. It has been observed however that, in some cases and situations, the concentrations of some trace elements (generally toxic and non-essential trace elements) in drinking water found in certain regions of a country could be significantly high so as to form the major fraction of the dietary trace element intake. High concentrations of Th and U in drinking water in the Monazite area located in southern region of India is one such example. The drinking water in this area contributed substantially to the daily intake of these elements [28]. The total area with such high concentrations of Th and U in drinking water, however, formed only a miniscule part of the total area of the country. This aspect was dealt with in depth during the RCM in the Philippines in July 1997 [29]. The participants agreed to treat such samples as a special case and not to include them in the main study.

#### 4.2.3. Problems faced in organ data collection in Bangladesh, Indonesia and Pakistan

All countries participating in the CRP did not agree to obtain organ content data on trace elements. In Bangladesh, Pakistan and Indonesia, due to cultural and religious constraints, autopsies are generally not permitted. Therefore, analytical data for organs could be obtained only from India, China, Republic of Korea and a limited amount from the Philippines. Japan already had some data available, hence new samples were not collected. Vietnam faced some logistical problems in sample collection and opted not to do so.

#### 4.2.4. Extraneous contamination

Diet and tissue samples can easily be exposed to external contamination leading to erroneous results. To avoid the contamination of samples with minerals and other trace elements during sampling and sample processing, the use of good laboratory practices and correct sampling and sample processing tools were adopted. The use of specified mineral and trace metal free materials such as Teflon knives and homogenizers fitted with titanium-coated blades and powder-free plastic gloves were recommended to the participants. The samples were homogenized after collection using a blender fitted with a titanium-coated blade. Some detailed studies on food processor erosion using ordinary stainless steel blades showed indications of erosion while processing hard materials.

#### 4.2.5. Calorie testing of the diet sampling method

The data sheet (Table 3), which the participants completed as the diet record, gives an idea of the total calorie content of diet sample representing the dietary intake in a particular country. From the known gross calorie (energy) intake of the participating countries, the investigation into the sampling methodology could be carried out in the case of a disagreement between reported and calculated calorie consumption from the collected diet. It was observed by Japanese workers at the CRL that the calorie consumption estimated from the duplicate diet in comparison with the model and total diet gave a lower estimate. This was useful when testing the representative food consumption in any country on the basis of the calorie consumption.

#### 4.3. Collection of human tissue samples

In addition to food samples, human tissue samples (both soft and hard tissue such as bone) were collected for analysis of trace elements. Complete details of tissue samples collected in various countries and elements determined in them are listed in Table 8. It is important to note possible errors that may arise while collecting tissue samples. Sometimes sampling errors imparted could be so significant that the sample quality is compromised beyond reasonable limits and thus render the

sample invalid for analysis [30]. The main problem in sampling the blood rich organs such as liver and kidney was to minimize residual blood. For these organs a quick rinse with high purity water to remove the bulk of blood or blotting with filter papers was adopted as a practical solution. Any further handling of samples beyond the sample collection in the autopsy room was avoided. It was considered imperative to transport the organs without much handling to the laboratory for removing the adipose and connective tissue as well as the blood and blood vessels. The organ by organ details of the protocols of sampling and sample handling procedures employed during collection are given below.

TABLE 8. ELEMENTS DETERMINED IN ORGAN CONTENTS STUDIES IN VARIOUS ASIAN	
COUNTRIES	
	-

Country	Human tissue analyzed					
	Lung	Liver	Kidney	Bone	S. Muscle	Thyroid
CHINA	Ca Cs K Sr Th U Cu Fe Mg Mn Na Se Zn Cd Hg Pb	Ca Cs K Sr Th U Cu Fe Mg Mn Na Se Zn Cd Hg Pb	Ca Cs K Sr Th U Cu Fe Mg Mn Na Se Zn Cd Hg Pb	Ca Cs K Sr Th U Cu Fe Mg Mn Na Se Zn Cd Hg Pb	Ca Cs K Sr Th U Cu Fe Mg Mn Na Se Zn Cd Hg Pb	Ι
INDIA	Th U	Th U	U	Sr Th U	Cs K Sr	Ι
REPUBLIC OF KOREA	Cs Sr Th U	Na K Cl Cs Sr I Th U				
PHILIPPINES	Cs Th U	Cs Sr Th U	-	-	-	-

#### 4.3.1. Liver

For handling liver samples, accessories made of Teflon were used to avoid extraneous contamination. The use of dust free vinyl gloves, pre-cleaned dust free Teflon sheets, containers including high purity water and titanium knives were considered mandatory. Liver samples were sealed in Teflon bags, frozen in liquid nitrogen and sent to the country laboratory.

#### 4.3.2. Kidney

Characteristics and also the elemental contents of the sample could get affected depending upon whether the tissue sample was collected from the cortex region or medulla or the distal part of a pyramid. It was therefore agreed by country laboratories to either use small samples from all the portions or collect the full kidney for later homogenization and analysis.

#### 4.3.3. Muscle

The skeletal muscle consists of long cylindrical muscle fibres varying in length from 1 to 40 mm and the thickness of up to 0.1 mm. Although there was not much problem in the sampling of this tissue, the only precaution taken was the consistency in the sampling site (which could be 2-3 in the body) for all the subjects.

#### 4.3.4. Bone

Bone is the mineralised matrix and comprises of bone tissue, red and yellow marrow, cartilage, periosteum and blood. They are generally of two types, the hard (compact) and the spongy trabaecular

bone. The sampling possibility is generally offered by 26 vertebrae and 25 ribs. The two to three sites chosen for sampling purposes were kept the same for all subjects.

#### 4.3.5. Thyroid

The thyroid gland weighs about 15–20 g and consists of the two lobes. The whole gland was removed, homogenized and taken up for analysis.

#### 4.3.6. Lungs

While collecting samples from lungs, a close examination of the lung walls for the heavy dust clusters was carried out and then avoided. It was also considered very important to avoid at all costs the portion of the lungs with pulmonary lymph nodes. Lymph nodes concentrations of Th and U are more than an order of magnitude higher than rest of the lung tissue [31] and could cause significant errors in results rendering their interpretation meaningless.

#### 4.4. Precaution during storage of samples

The validity of a sample when stored for longer duration is generally of concern since the studies on matrix stability (especially with respect to organic species) following extended periods of preservation are scarce. The results of the extensive investigations that were carried out at the National Institute of Standards and Technology (NIST), USA to evaluate storage stability, showed no significant changes for Se and Zn. Some loss of As from bovine liver matrix, however, was suspected with prolonged storage [32]. Similarly, human specimens of blood and liver were shown to be stable to Cd and Pb [33] during fairly long periods of storage but some doubt, however, exists on the behaviour of volatile elements such as Hg and As during storage. The lyophilised human body fluid reference materials, however, were shown to be stable for a period of 5 years for Hg, Pb and Al [34]. In the present situation, the study was mainly for trace elements where the storage of samples free of extraneous contamination was the only key requirement.

Fresh tissue and diet composite slurries were frozen in Teflon bags or high-density polyethylene bottles and preserved at  $-20^{\circ}$ C. When long term storage was anticipated, the material was freeze dried prior to analysis.

Changes in concentrations of elements due to precipitation, absorption and evaporation of the aqueous medium were prevented until measurement was complete. High density PE-bottles were found acceptable for short term storage and the long term storage was carried out in Teflon/quartz containers.

#### 4.5. Sample processing

Much progress has been made in the trace element analysis of biological samples with enhanced knowledge in sampling, sample preparation, instrumentation, awareness in the matrix interference and preparation of primary standards. The analytical standardization process, however, still suffers from the problem of either the increase or loss in the analyte due to either contamination or the absorption process. Whereas the contamination is the result of chemical processing of the sample, the loss of the analyte is often due to the matrix dissolution problem leading to incomplete recovery. At times the loss could also be due to volatilisation of some elements. Therefore, precautions were taken while processing the samples to avoid the contamination or loss, using the guidelines described below.

#### 4.5.1. General guidelines

For processing of samples which included, freeze drying, ashing, powdering, dissolution, sealing of samples (in case of NAA technique) for their transportation to analytical laboratory prior to analysis, following general guidelines were provided by the CRL to various country laboratories and were also implemented.

#### 4.5.2. Special homogenizers fitted with Ti-coated blades

Mixer blenders fitted with the ordinary steel blades could contain trace elements Co, Mn, Cr, *etc.* and contaminate the homogenized samples. Some detailed studies on food processor erosion using ordinary stainless steel blades have been carried out at University of Massachusetts (USA), which showed no erosion when soft material was processed but there were indications of erosion while processing the hard materials [35]. Hence, in order to overcome this problem participating laboratories received special blenders fitted with Ti-coated blades obtained through the assistance of NIRS, Japan (CRL) and supplied by IAEA.

#### *4.5.3. Laboratory environment*

The laminar flow clean hood was used wherever possible for carrying out various activities connected with the sample preparation. Proper segregation of the laboratory space for different uses was also carried out and provision for clean conditions was made mandatory to prevent inadvertent contamination of samples during processing.

#### 4.5.4. Procedures for non-destructive methods

As far as possible, homogenized samples were freeze dried. The ashing of the samples was avoided to ensure there was no loss of volatile trace elements such as iodine and also to some extent selenium.

#### 4.5.5. Ashing and dissolution of the sample

No special ashing apparatus was required for elements of importance in radiological protection with the exception of iodine. Precautions were taken to avoid the contamination of samples from inner walls of the muffle furnace. The maximum recommended temperature of 450<sup>°</sup>C was allowed to reach slowly when dealing with soft tissues and also food samples. Care was taken to recover elements from the ash quantitatively, ensuring dissolution by applying proper acid leaching or treatment in a PTF beaker with HF, following the decomposition of the organic substance until clear solution was obtained. This step ensured reliable results.

#### 4.5.6. Matrix problem

The INAA was used for measuring total quantity of trace elements in material before and after dissolution in order to measure any loss of analytes associated with the measurement of trace elements in plant materials containing appreciable amounts of silica. This kind of problem was relevant to the measurement of Th and U concentration in diet.

#### 4.6. Tissue sample processing

#### 4.6.1. Preparation of tissue samples for different analytical methods

The main analytical techniques employed in the CRP for the analysis of tissue samples were INAA, RNAA, ICP-MS, ICP-AES and, in a few cases, AAS. Whereas no sample dissolution was required when using INAA, the sample needed to be dissolved or brought into solution for the other three techniques. Certain precautions were required to be taken during dissolution when employing any one of the three methods. Considerable guidance available from the published literature was utilized [37].

#### 4.6.2. Dust-free environment

The concentrations of most of the trace elements were generally low in tissue samples compared to those present in diet samples with the exception of the few trace elements such as I in thyroid. Precautions stated above regarding the handling of diet samples were therefore followed more strictly.

#### 4.6.3. Homogenization and freeze drying

Tissue samples were collected from as many different sites of the organ as possible and then cut into small pieces with the help of titanium coated blade and finally homogenized with the help of the mixer blender fitted with titanium coated blade before freeze drying and final powdering. Most other steps required for processing the diet samples were valid for processing tissue samples as well.

#### 4.6.4. Processing of samples for NAA

Tissue samples were freeze dried and then ground in a clean agate mortar. For NAA, the required amount of tissue or food samples (typically between 50-250 mg) was heat-sealed in a pre-cleaned polyethylene bag and double bagged with the identification tag in the outer bag which, after irradiation, was discarded. The inner bag after insertion into a new bag was re-identified and taken for radioactivity measurement. In the case of neutron irradiation, when the total fluence exceeded  $10^{17}$  neutrons, the quartz capsule was used for irradiation of the sample instead of the polyethylene bag.

#### 4.6.5. Wet digestion

The classical open system consisting of a borosilicate beaker with watch glass for cover was found adequate and used in the wet digestion of the biological samples. Closed systems such as Teflon pressure vessel and metal jacket for acid decomposition were considered effective for trace and other elements for good performance and were used wherever available. An automatic microwave oven system provided which improved efficiency was also used by some participants at the CRL.

#### 5. ANALYTICAL METHODOLOGY

Concentrations of Cs, Sr, I, Th and U in diets and human tissues were so low (sub-nanogram) that the existing analytical methods were not adequate to quantify them. There were practically no CRMs available for the validation of available analytical methods. The first challenge therefore faced by the CRP participants was to develop and standardize methods capable of measuring concentrations of these five trace elements in diet and human tissue matrices. The next step was to validate them by using CRMs with biological matrices similar to those encountered in human diet and tissue materials.

The ability of some currently employed modern analytical techniques to determine the concentrations of Cs, I, Sr, Th and U in diet and human tissue samples is already shown in Tables 5A–5E. It is clear from those tables that whereas AAS and INAA are capable of measuring one or two elements, ICP-MS, RNAA and ICP-AES could effectively measure concentrations of all the elements of interest both in tissue and diet samples. On the other hand, AAS is effective for the determination of minor elements Na, K, Ca, Mg and trace elements Cu, Zn, Mn and Fe. INAA is useful for determining Cs, Cr, Se, (and RNAA for I) in addition to elements that can be analysed by AAS. In a recent publication, Akhter *et al.* have reported the feasibility of using INAA for measuring Th in diet [36]. The concentrations measured by them in diet samples were however higher than those encountered in diet samples in many other Asian countries.

As part of this CRP, RNAA and ICP-MS were developed and standardized for most elements and AAS was used for Ca and Sr. RNAA was developed in the country laboratory in India for measuring Th, U, Sr and I in diet and tissue samples. The same method was also used for the analysis of samples from Republic of Korea. Analytical methods based on ICP-MS and ICP-AES were successfully standardized and applied by the CRL (NIRS, Japan) for determining Th, U, Ca, Cs and also some other elements in diet and tissue samples.

The expertise developed for determining Th, U, Sr and I, using RNAA in India, was transferred to the country laboratory at the Republic of Korea under an IAEA expert exchange programme. One participant from the Philippines and two from Vietnam were trained at the CRL, Japan, in the application of ICP-MS for the analysis of diet and tissue samples.

Details of analytical methods developed by participating countries for the analysis of food and tissue samples for various elements are discussed under 5.1.

The details of the characteristic gamma ray energies of the isotopes of interest for the determination of Th, U, Sr, I and also for a number of other minor, essential and toxic trace elements are included in Table 9. Also shown in the Table are the emission lines for some of the elements analysed by AAS and ICP-AES.

#### 5.1. RNAA for determining concentrations of Th, U, Sr and I in diet and tissue samples

#### 5.1.1. Thorium

Th in biological samples was determined using RNAA by measuring the activity of <sup>233</sup>Pa, produced on irradiation of Th in the sample. The nuclear reaction taking place is shown below:

$$^{232}$$
Th (n,  $\gamma$ )  $^{233}$ Th-----  $\beta^{-------}$   $^{233}$ Pa

<sup>233</sup>Pa activity in the irradiated sample was radio-chemically separated from other interfering activities, and counted using the 311.8 keV characteristic gamma ray.

Mn carrier of 12.5 mg/mL concentration was prepared from manganese dioxide by dissolving it in nitric acid in the presence of 30% hydrogen peroxide. 4M nitric acid was prepared by appropriately diluting the concentrated nitric acid.

Element	Isotope of interest	Half-life (d)	Gamma energy of interest (keV)	Emission line (ICP-AES)
Са	<sup>47</sup> Sc	3.6	160	315.9
Cs	<sup>124</sup> Cs	754.2	795	-
Ι	<sup>128</sup> I	0.174	442	-
K	<sup>42</sup> K	0.515	1524.6	766.5
Sr	<sup>87m</sup> Sr	0.117	389	407.8
Th	<sup>233</sup> Pa	27.0	311.8	401.9
U	<sup>239</sup> Np	2.36	227.5 & 278	385.9
Cd	<sup>115</sup> Cd	2.23	528	-
Co	<sup>60</sup> Co	1924	1173 & 1332.4	231.2
Cr	<sup>51</sup> Cr	27.8d	320	205.6
Fe	<sup>59</sup> Fe	45.6	1098.6 & 1291.5	238.2
Hg	<sup>203</sup> Hg	46.9	279	253.7
Mg	<sup>27</sup> Mg	0.06	844	279.8
Mn	<sup>56</sup> Mn	0.104	857	257.6
Na	<sup>24</sup> Na	0.625	1375 & 2750	589.6
Se	<sup>75</sup> Se	127	136 &265	196.0
Zn	<sup>65</sup> Zn	245	1115	213.8

TABLE 9. NUCLEAR PARAMETERS AND EMISSION LINES FOR MEASURING SELECTED TRACE ELEMENTS WHEN USING NAA, OR ICP-AES

The sample containing <sup>233</sup>Pa was allowed to cool for 10–15 days for short-lived interfering activities of <sup>24</sup>Na, <sup>42</sup>K, and <sup>38</sup>Cl to decay. Subsequently the sample was dissolved in concentrated nitric acid with occasional addition of a few drops of hydrogen peroxide until clear solution was obtained. 50 mg Mn carrier (4mL of carrier solution) was added and the solution heated to near dryness. 25 mL of 4M nitric acid followed by 150–200 mg of potassium bromate and the solution was heated gently to obtain black precipitate of manganese dioxide. This manganese dioxide precipitate quantitatively carried the <sup>233</sup>Pa. The precipitate was centrifuged and the supernatant was discarded. It was then dissolved by the addition of a few drops of nitric acid and also one to two drops of hydrogen peroxide. Seven mL of concentrated sulphuric acid and 200 mg of potassium sulphate were added to the solution. It was heated gently and 20 mL of barium carrier (4 mL of barium chloride solution of 5 mg/mL Ba) was added drop wise to precipitate barium sulphate.

The solution containing barium sulphate precipitate was heated until the release of sulphur trioxide fumes when precipitate was completely re-dissolved. This solution was cooled and 40–50 mL of water were added to it with constant stirring. The barium sulphate in the solution got re-precipitated. To this, additional 4 mL barium carrier solution was added drop-wise. The two steps ensured quantitative co-precipitation of <sup>233</sup>Pa along with barium sulphate precipitate. The precipitate was allowed to settle for 2–3 hour and then filtered using Whatman No. 42 filter paper, dried and counted for 311.8 keV gamma ray of <sup>233</sup>Pa, using hyper-pure germanium detector coupled to multi-channel analyser (MCA). The irradiated standard of Th was also chemically processed in the same manner and then counted on the same detector and in the same counting geometry. The quantification of Th was carried out by comparing the activities of <sup>233</sup>Pa in sample and the standard.

#### 5.1.2. Uranium

When using RNAA, U in sample could be determined by measuring <sup>239</sup>Np, which is produced on neutron irradiation of U present in the sample. Following nuclear reaction takes place:

$$^{238}$$
U (n,  $\gamma$ )  $^{239}$ U ------  $\beta^{-}$  ----- $^{239}$ Np

The <sup>239</sup>Np has a half-life of 2.33 days and emits characteristic gamma ray of 228 keV and 277.5 keV. The quantification of U in sample was carried out by radiochemical separation of <sup>239</sup>Np from it and measuring its activity and comparing it with that induced in the known amount of U standard. The details of the radiochemical separation are given below.

The irradiated food sample was digested in about 10 mL of concentrated nitric acid and 1–2 mL of 30% hydrogen peroxide was added drop wise to complete the digestion. A known quantity of <sup>238</sup>Np activity (obtained on irradiation of a few nanogram of <sup>237</sup>Np, irradiated along with the sample) was also added while digesting the sample. On completion of digestion, the solution was heated until almost dry. Thirty mL of 9M HCl was added to it, along with 0.5 g hydroxylamine hydrochloride, while continuing to gently heat it for ten minutes. This step ensured the conversion of all Np present in the solution to Np+4 state.

About 2g of DOWEX 1X8 (100–200 mesh size) anion resin in the chloride form was taken in a beaker containing distilled water and allowed to swell to its maximum size. The resin was loaded on to an ion exchange column. The column dimensions were about 1cm (diameter) and 12 cm (ht.). The solution containing <sup>238&239</sup>Np was then passed through the column which was previously equilibrated with 9M HC1. The flow rate was adjusted to 0.5 mL per minute. The column was washed with two 30 mL aliquots each of 9M HCl to allow interfering activities to be washed down, while <sup>239</sup>Np and <sup>238</sup>Np were retained. Both isotopes of Np were eluted from the column with 60 mL of 1M HCl.

Seven mL of sulphuric acid and 200 mg of potassium sulphate were added to the solution containing <sup>238 & 239</sup> Np-activity and barium sulphate was precipitated from it by the drop wise addition of barium carrier (as barium chloride solution of 5 mg /mL). The precipitate of barium sulphate which quantitatively carried Np along with it, was allowed for 2 hours to settle down and then filtered through Whatman No. 42 filter paper. The precipitate was then dried, mounted and counted using 54 CC hyper-pure Germanium detector coupled to a 4K MCA. The chemical yield recovery correction for <sup>239</sup>Np was calculated by measuring the activity of <sup>238</sup>Np- before and after the completion of radiochemical procedure. The characteristic gamma ray of 984 keV emanating from <sup>238</sup>Np was employed for this purpose.

#### 5.1.3. Strontium

Strontium was measured in biological samples using RNAA, by separating <sup>87m</sup>Sr produced from it on neutron irradiation when <sup>87m</sup>Sr is formed by the following nuclear reaction:

$${}^{86}$$
Sr (n,  $\gamma$ )  ${}^{87m}$ Sr

<sup>87m</sup>Sr has a half-life of 2.83 h and emits a characteristic gamma ray of 389 keV. The radiochemical separation of <sup>87m</sup>Sr from the sample is quantitative (>95%) and there is no need to apply chemical recovery correction. It is however possible to determine the chemical recovery factor by using long-lived <sup>85</sup>Sr activity. The details of the radiochemical separation procedure are given below.
Sample was digested in concentrated nitric acid in the presence of about 100  $\mu$ g of (stable isotope) strontium, until clear solution was obtained. About 1–2 mL of 30% hydrogen peroxide was used to aid digestion and to the digested sample about 50 mg calcium carrier (as calcium chloride) was added to ensure that pH of solution is acidic. 2–3 mL of concentrated hydrochloric acid was added followed by 3–5 mL of 5% ammonium oxalate. The pH of the solution was raised between 2 and 3 by drop wise addition of ammonium hydroxide to precipitate calcium oxalate from the solution. It quantitatively carried <sup>87m</sup>Sr along with it.

There was no need to determine chemical recovery since Sr gets carried on to calcium oxalate quantitatively. However, in the initial stages of standardizing the method, <sup>85</sup>Sr was used as carrier and the yield correction factor was obtained by measuring characteristic gamma ray of 514 keV of <sup>85</sup>Sr. The precipitate was filtered using Whatman No. 42 filter paper and counted for characteristic gamma ray of 389 keV. The irradiated Sr standard was also subjected to the same radiochemical separation and counted using 54 CC germanium detector coupled to MCA in the same geometry. Strontium in the sample was quantified by comparing <sup>87m</sup>Sr activities in sample and the standard.

# 5.1.4. Iodine

The sample containing iodine was digested in a microwave oven in a closed system in 1-2 mL of ultra-pure nitric acid. After digestion, the system was opened after cooling it to about  $-40^{\circ}$ C. To the clear solution of digested sample, I-131 tracer (carrier free) was added along with bismuth carrier (about 20 mg as bismuth nitrate). Iodine present in the sample was reduced to iodide, using hydrazine hydra-sulphate. The pH of the solution was adjusted between 2–3 by adding a few drops of ammonia to it. The iodine was then co-precipitated with bismuth sulphide using thioacetamide. The chemical yield was determined by measuring characteristic gamma ray of 362 keV of known activity of I-131 added after the digestion stage. The precipitate, containing I was filtered, dried, sealed and irradiated for 5 minutes in a flux of  $10^{12}$  neutrons cm<sup>-2</sup>. sec<sup>-1</sup>. in a swimming pool type of reactor, when the stable iodine got converted to <sup>128</sup>I by the following nuclear reaction:

$$^{27}$$
I (n,  $\gamma$ )  $^{128}$ I

1

After irradiation, the precipitate was counted using the 442 keV gamma ray of <sup>128</sup>I. Iodine was quantified by counting and comparing activities produced in sample and known standard.

The analytical methods developed and standardized were further validated by the analysis of three CRMs procured from NIST (USA). These analytical methods were then used for generating reference values for three other RMs from NIST namely, total diet, typical diet and bone meal along with the certification of the Typical Japanese Diet RM prepared by NIRS in collaboration with the National Institute for Environmental Studies (NIES), Japan.

The results of the analysis of NIST SRMs, namely, citrus leaves, orchard leaves and the bovine liver and also of typical Japanese diet, for the above three elements, are shown in Table 10.

# 5.2. Determination of iodine in diet samples at a contract laboratory

Due to its low concentration in diet samples and because of its volatile nature, iodine is considered a difficult element to determine. Most of the participating country laboratories did not have adequate analytical capabilities to determine its concentration in diet samples. It was therefore determined as part of a separate IAEA research contract given to Nuclear Physics Institute, Rež near Prague, Czech Republic. Iodine was determined in diet samples collected in the Asian countries participating in the CRP, using both epithermal neutron activation analysis (ENAA) and RNAA. The choice of the method depended on the iodine level and also upon the level of other interfering elements which hampered its non-destructive determination by ENAA. In order to ensure reliability of the analytical method several RMs were analysed for quality control. Both the analytical methods are described below.

TABLE 10. COMPARISON OF THE RESULTS FOR CRMS AND TYPICAL JAPANESE DIET RM WITH THE CERTIFIED VALUES.USING THE ANALYTICAL METHODS DEVELOPED FOR CRP

Element	Citrus Leaves		Orchard Leaves		Bovine Liver		Typical Japanese Diet RM	
	Present work	Certified Value	Present work	Certified Value	Present work	Certified Value	Present work	Certified Value
I (µg/g)	1.80 <u>+</u> 0.1	1.84 <u>+</u> 0.03	0.20 <u>+</u> 0.05	(0.17)	N.D.	(0.2)	1.73 <u>+</u> 0.2	1.9
Sr (µg/g)	95 <u>+</u> 4	100 <u>+</u> 2	41.0 <u>+</u> 2.5	37.1 <u>+</u> 1	0.14 <u>+</u> 0.01	0.14	4.7 <u>+</u> 0.2	4.9 <u>+</u> 0.2
Th (ng/g)	13.0 <u>+</u> 2.0	15.0 <u>+</u> 2.0	60.0 <u>+</u> 2.8	64 <u>+</u> 6	3.9 <u>+</u> 0.2	3.2 <u>+</u> 0.2	2.4 <u>+</u> 0.06	2.0-4.6 (*)
U (ng/g)	32.0 <u>+</u> 2.0	33.0 <u>+</u> 4.0	27.0 <u>+</u> 2.0	29 <u>+</u> 5	0.7 <u>+</u> 0.1	0.8 <u>+</u> 0.1	3.15 <u>+</u> 0.08	2.9 <u>+</u> 0.4

(\*) Range of recommended values

#### 5.2.1. ENAA

Samples weighing approximately 100–120 mg were heat sealed in acid-cleaned polyethylene (PE) capsules and irradiated in 1-mm thick cadmium vials for 30s in a nuclear reactor LVR-15 of the Nuclear Research Institute Rez, operated at 9 MW. After a decay time of 15 to 20 minutes, samples were counted for 20 to 25 minutes using a coaxial HpGe detector (relative efficiency 23%, resolution FWHM 1.8 keV, for 1332.5 keV photons of <sup>60</sup>Co).

#### 5.2.2. RNAA

Samples weighing 130–150 mg were irradiated in the reactor for 2 minutes and subjected to a radiochemical separation procedure based on alkaline fusion and extraction of elementary iodine in  $CHCl_3$  that has been described in detail elsewhere [38]. After 16 to 18 minutes of separation, the iodine fraction was counted as described above. The chemical yield was in the range 90 to 95% as ascertained using <sup>131</sup>I as a radiotracer in each experiment.

#### 5.2.3. Validation of analytical methods for iodine

As part of the quality control to ensure the generation of the reliable results, CRMs obtained from NIST (USA) were analysed for iodine using methods standardized for the analysis of iodine. Table 11 compares the results obtained for the NIST RMs and SRM, respectively with NIST certified (with uncertainties), non-certified or literature values. Iodine levels > 100 ng g<sup>-1</sup> were mostly determined by ENAA, while for the determination of lower iodine levels, RNAA was used. The results agreed with the NIST certified values wherever available, within the uncertainty margins. The measure of uncertainty of results was a combined uncertainty (1s), in which the standard uncertainties of the preparation of standards, irradiation and counting parameters such as the counting statistics were incorporated [39]. The dispersion of individual results was also considered. Obviously, this testified the accuracy of ENAA and RNAA procedures. The results also agreed with those reported for some RM and SRM analysed recently [40]. The results for RMs were reported on dry mass basis and were determined according to instructions given in the respective certificates or non-analysed aliquots.

### 5.3. Determination of Sr in biological samples using AAS

#### 5.3.1. Sr in diet

A special method for the determination of Sr in food samples using AAS was developed at the CRL, NIRS (Japan). The analytical details of the method are described below.

TABLE 11. QUALITY CONTROL IN THE ANALYSIS OF ASIAN DIET: RESULTS FOR IODINE DETERMINATION IN US NIST RMs BY INAA AND RNAA (in ng/g dry mass unless otherwise stated)

Reference Material	ENAA	RNAA	Mean <u>+</u> unc.	NIST value	Literature value
SRM-1575		87.8, 87.8 90.0	88.2 <u>+</u> 5.3	-	80 <u>+</u> 30
Pine needles		92.8,88.1,83.5			235 <u>+</u> 15
		82.8,92.9			145
SRM-1571		210, 205, 205	207 <u>+</u> 6	170	155 <u>+</u> 10
Orchard Leaves					186 <u>+</u> 18
RM-8433		31.1, 26.8, 24.4	27.1 <u>+</u> 4.0	26 <u>+ 6</u>	-
Corn Bran		26.0			
RM-8414	<45	43.0, 40.9, 39.8,	39.9 <u>+</u> 3.3	35 <u>+</u> 12	40 <u>+</u> 20
Bovine Muscle		43.4, 35.0, 37.1			
SRM-1486	112, 99, 130, 118	96.7, 104.6	110 <u>+</u> 22	-	110 <u>+</u> 40
Bone Meal					
SRM-1548	400, 399, 426,	376, 388	394 <u>+</u> 42	-	350 <u>+</u> 110
Total Diet	372				
SRM-1548a	657, 676, 711,	685	713 <u>+</u> 57	759 <u>+</u> 103	600 <u>+</u> 200
Typical-Diet	805				
SRM-1566a (µg/g)	4.68, 4.71, 4.85	4.74	4.74 <u>+</u> 0.16	4.46 <u>+</u> 0.42	4.3 <u>+</u> 0.4
Oyster Tissue					

The ashed food sample was taken along with 30 mg carrier of Ca and concentrated hydrochloric acid, and heated to dryness in a water bath. It was then dissolved in demineralised water and filtered. Any precipitate containing silica was discarded. The silica free filtrate was gently heated along with the addition of 5 mL of 5% ammonium oxalate and pH was adjusted to 4.6. The solution was heated gently to coagulate calcium oxalate precipitate which quantitatively carried all the Sr. The precipitate was filtered and filtrate discarded. The calcium oxalate precipitate was ashed at about 550°C for several hours and then dissolved in a few mL of dilute nitric acid. It was heated to complete dryness and residue was taken up in 0.1 M HCl. Sr in the sample was determined employing normal AAS procedure by using characteristic absorption line of 460.7 nm.

### 5.3.2. Determination of Ca and Sr in bone ash

The connecting tissues were removed from the bone sample and it was first dried at  $110^{\circ}$ C and subsequently ashed in an electric muffle furnace by increasing the temperature stepwise to 550°C. This temperature was maintained for at least 12 hours. The bone sample was then pulverised using agate mortar and pestle. The powdered bone sample was stored in a plastic bag and an aliquot weighing about 400 mg was taken up for analysis. Five mL of 1M HCl was added to dissolve the sample and then further diluted to 50 mL (the bone ash solution concentration was 0.1M HCl).

An anion exchange column (Dowex 1x8, 100–200 mesh size) of the size  $2 \text{ cm} \times 25 \text{ cm}$  was fitted and preconditioned with 0.1 M HCl solution. The solution was passed through the column and the first 2/3 of the eluate was discarded. The second 1/3 of the phosphate free eluate was then collected in a test tube. 2 mL of this phosphate free bone solution was taken in a 50 mL beaker and 1 mL of triethanolamine was added to mask Fe, then NN indicator for complexo-metry and 5 mL of KOH solution was added to make the solution alkaline. Finally the volume was made to 30 mL with demineralised water. The Ca was titrated with 0.02m EDTA standard solution for the colour to turn pink from blue. 12 mL (or 2/3 of the solution) of the phosphate free bone solution was taken for the measurement of Sr using AAS using the characteristic absorption line of 460.7 nm.

#### 5.4. Determination of trace elements by ICP-MS

The main analytical methods employed by the CRL at NIRS Japan for the analysis of diet and tissue samples were ICP-MS and ICP-AES. The CRL was responsible for the external quality control and as back up laboratory for countries where expertise for the analysis of some elements did not exist. It also carried out the analysis of all samples of diet materials as part of the Japanese data reported for the CRP. The description of methodology adopted for sample preparation for the two analytical methods employed by the CRL is given below.

The samples were chemically treated in a Class 100 clean air draft chamber installed in a Class 10 000 or Class 5000 clean room (switched to the latter in May 1999). For sample dissolution a programmable microwave digestion apparatus was used together with Tama Pure AA-100 ultra-pure nitric, hydrochloric, and perchloric acids and hydrogen fluoride. A Barnstead Nanopure and Nanopure Infinity ion exchange apparatus was used to produce purified water (switched to the latter in May 1999). Decomposition vessels and Teflon beakers were first washed using detergent, then soaked in warm dilute nitric acid overnight, rinsed with purified water, dried in a clean-air hood and stored in polyethylene bags until use. Disposable plastic gloves for clean room work and a mask with charcoal filter were used to prevent possible contamination from fingers and also to avoid the health problems due to acid fumes.

Determination of Cs, Sr, Th and U was carried out by ICP-MS; Ca, K, Mg, Fe, Zn, Cu and other essential elements by ICP-AES. The CRL did not use AAS because of its time-consuming nature during the present phase of the RAM- CRP.

The sample solution was diluted 2.5 and 2,000 fold with 10% HNO<sub>3</sub> shortly before the elemental measurement with the addition of Rh and Bi as internal standards for Sr, Cs and Th, U respectively. Analytical curves were prepared from commercially available spectrometric standard solutions by diluting them 3 or 5 times in concentration levels, and also adding the internal standards.

An ICP-MS instrument was used under the following conditions: ICP plasma torch with microwave frequency 27.12 MHz and output 1.2 kW, position of ion sampling 7.5 mm, sample uptake rate 0.4 mL/min., total Ar gas flow rate 16.9L/min., scanning mass range 88–133 and 209–239 m/z for <sup>88</sup>Sr, <sup>133</sup>Cs, and <sup>232</sup>Th, <sup>238</sup>U respectively. The number of measurement points per peak was 3, number of scan sweeps 100, dwell time per point 33 ms, integration time 3.3 s, overall triplicate measurements per sample being 100 s.

# 6. QUALITY ASSURANCE (QA)

One significant challenge in this CRP was to generate reliable and accurate data on Cs, I, Sr, Th, U, etc. which were present at such low concentrations that the samples could easily get contaminated or the concentrations could get depleted due to adsorption of these elements on to the container walls. There were hardly any CRMs available to allow participating laboratories to carry out internal quality control for the data generated. In order to meet these challenges, the IAEA took the first step by providing the participants with RMs of known trace element concentrations.

With the objective of acquiring improved reference values for the above stated elements, a coordinated study was undertaken by the IAEA in which four SRMs: SRM-1548 Total Diet, SRM-1548a Typical Diet, SRM-1486 Bone Meal and SRM-8414 Bovine Muscle supplied by NIST were distributed to seven selected laboratories in five countries. In these SRMs, all the five elements were analysed with the help of highly sensitive analytical techniques like INAA, RNAA, ICP-MS, etc. These laboratories were selected on the basis of their known expertise in the kind of analyses required. The data was collected and evaluated by the CRL at the NIRS. Two additional RMs, NIST-SRM-1575 Pine Needles and NIST-SRM-1566a Oyster tissue, were also used since they had the advantage of having a few certified elements in them. Pine Needle was certified for Cs, Th and U and Oyster Tissue certified for I, Sr and U. Thus, these two materials covered all the five elements whose concentrations were sought to be certified. The analysis of these two materials gave confidence to analytical laboratories involved in the certification process.

A summary of the results obtained for six materials and five elements is given in Table 12. Wherever certified values were available, a good agreement with certified values was seen. The tentative reference values assigned by this study were sufficiently accurate to be used for the purpose of the CRP. It may be stated here that although the analyses of all these materials was attempted for all elements, results are not needed for all these materials. For example, there was no need to assign certified values other than those of Th, U and Sr in bone. In the same way, there was no need to certify the concentration of iodine in any tissue other than thyroid.

# 6.1. Internal Quality Control

Once the four RMs were assigned concentration values for the five elements of interest, they were distributed to the nine participating laboratories to validate analytical methods developed by them. The participants were advised to use these materials to validate and also to run one RM along with every tenth real sample analysed so that a continuous internal check could be carried out on the data being produced in the laboratory. They were also encouraged to use, for quality control, any additional RMs available with them which had certified values for the elements of interest.

#### Central reference Laboratory (CRL) Concept

The hallmark of this CRP was implementation of strict quality control both internal and external. Whereas internal quality control was ensured by validation of analytical methods within the country laboratory, the external quality control on the other hand was assured by appointing the participating laboratory at NIRS, Japan, as the CRL. This laboratory was assigned the following responsibilities:

#### 6.1.1. The role of CRL

- On request from other participants, to provide guidance, training and support in various matters dealing with the CRP such as sample preparation, storage, analysis, etc.;
- To analyse a representative number (10%) of samples collected and analysed by other participants in order to provide an independent cross-checking of data for external quality control purposes;

- To assist CRP participants with analytical quality assurance by developing reference values for some selected NIST RMs which had not previously been certified for the elements of interest in this CRP;
- To developed a new reference material with similar matrix as the diet samples such as an Asian reference diet. The CRL produced the Typical Japanese Diet RM in collaboration with NIES and certified it for a large number of trace elements [41].
- To assist the IAEA with periodic external quality control exercises and to evaluate the data obtained, in part through the calculation of z-scores.
- To undertake a central compilation and evaluation of the data for selected high priority specimens and elements as reported by all participants in the CRP.

# TABLE 12: REFERENCE VALUES ASSIGNED FOR USE BY THE CRP PARTICIPANTS FOR THE RELEVANT ELEMENTS AND MATRICES [40]

Sample name	Element	Ν	Mean	S.D.	RSD(%)	Assigned reference value	Certified value
SRM 1486	Sr	16	264.5	16.2	6	265+32	264+7
Bone Meal	Cs	17	0.99	0.42	42	$1.0 \pm 0.8$	- <u>-</u> ·
	Th	23	4.65	0.83	18	4.7 +1.7	
	U	24	18.6	5.8	31	19 + 12	
	Ī	8	0.11	0.02	18	0.11 + 0.04	
SRM 1548	Sr	17	3.7	0.9	24	3.7 + 1.8	
Total Diet	Cs	27	13.7	2.5	18	14 + 5	
	Th	19	1.2	0.5	39	1.2 + 0.9	
	U	24	2.2	0.6	29	2.2 + 1.2	
	Ι	14	0.35	0.05	15	0.35 + 0.1	
SRM 1548a	Sr	13	3.3	0.4	13	3.3 + 0.8	
Typical Diet	Cs	22	10.0	1.6	16	10+3	
~ 1	Th	14	0.83	0.26	31	0.8 + 0.5	
	U	21	2.2	0.7	32	2.2 + 1.4	
	Ι	12	0.64	0.09	14	0.6 + 0.2	
SRM 1566a	Sr	17	11.4	1.3	12	11+3	11.1+1.0
Oyster Tissue	Cs	24	17.8	2.9	16	18 + 6	—
-	Th	23	37.7	4.8	13	38 + 10	
	U	25	128	12	9	128 + 24	132+12
	Ι	15	4.27	0.19	4	$4.31 \pm 0.4$	$4.46 \pm 0.42$
SRM 1575	Sr	17	47	0.8	17	4 7+1 6	4 8+0 2
Pine Needles	Cs	28	115	11	9	115+22	1.0_0.2
	Th	26	32.3	7.5	23	$32 \pm 15$	37.3
	U	25	15.4	2.9	19	15+6	20.4
	Ī	11	0.08	0.02	20	$0.08 \pm 0.31$	
SRM 8414	Sr	16	0.06	0.01	19	0.06 <u>+</u> 0.02	0.052 <u>+</u> 0.015
Bovine Muscle	Cs	28	34.0	4.2	12	34 <u>+</u> 8	—
	Th	20	1.00	0.89	89	(1.0)	
	U	16	0.26	0.14	54	(0.3)	
	Ι	11	0.038	0.012	31	0.04 <u>+</u> .02	0.035 <u>+</u> 0.012

Units: Cs, Th and U are in ng/g and I and Sr in ug/g.

### Internal Quality Control at the CRL (NIRS, Japan)

Since responsibility for quality control of the analyses carried out in the various country laboratories was entrusted to the CRL, it was important to validate the analytical methodologies developed at the CRL itself by analyzing certified reference materials.

A comparison of results of the analysis of RMs carried out at the CRL along with the certified values is shown in Table 13. Results reported by the CRL agreed very well (within 10%) with the certified values.

#### 6.1.2. Analytical contributions of the CRL

The CRL provided the overall quality control support. In addition, 460 determinations for different trace elements in food samples and 151 determinations on tissue samples were carried out (152 food and diet samples and 57 tissue samples). These samples of tissue and food were analysed for different Asian countries which did not have analytical expertise for one or more of the elements. This analytical contribution of the CRL was to ensure the generation of reliable data by the CRP participants.

CRM			Caesium			Thorium			Uranium		S	Strontiun	1
		µg /g	Mean	SD	µg/g	Mean	SD	µg /g	Mean	SD	μg /g	Mean	SD
NIST SRM 1575 (Pine needles)	1 2	0.1169 0.1166	0.117	0.0002	0.0392 0.0362	0.038	0.002	0.0184 0.0186	0.018		4.75 4.74	4.7	0.01
Certified Value		0.115		0.0022	0.037		0.003	0.020			4.8		0.2
NIST SRM 1548a (Typical Diet)	1 2 3	0.0098 0.0096 0.0098	0.0098	0.0001	0.0008 0.0009 0.0008	0.0008		0.0023 0.0023 0.0023	0.0023		2.923 3.088 3.009	3.01	0.08
Certified Value		0.0098		0.0003	0.0008		0.0005	0.0022		0.001	2.93		0.10
NIST SRM 1571 (Orchard Leaves)		0.039	0.039		0.060	0.060		0.030			36.45		
Certified Value		0.040		0.0004	0.064		0.005	0.029		0.005	37.0		1.0

# TABLE 13. QUALITY CONTROL RESULTS OF ANALYSIS CARRIED OUT AT THE CRL FOR Cs, Th, U AND Sr IN CRMs

#### 6.2. External quality control

It was agreed at the project formulation meeting [42] that, as part of the external quality control exercise, the duplicate aliquots of the 10% of the total food and tissue samples analysed in various country laboratories should be sent to the CRL for analysis. Special guidelines on preparation and shipment of samples to the CRL were prepared and distributed to the participating country laboratories and are shown in Table 14. The results of the analysis for those samples carried out at the country laboratory and at the CRL were compared and the standard z-score test [43] was applied to the data on various elements.

Typical sample	mixed diet
	autopsy specimen (bone, liver, etc.)
	selection of both should be randomized over the collection period
Amount to be supplied	Homogeneous dry powder
	7–10 g for food samples
	2–5 g for tissue samples
	Ash
	1–1.5 g (for all kinds of sample)s
Sample containment	Small plastic bottle or self-sealing polyethylene capsule
(either of the following)	
	clean inside and outside with ordinary washing water
	then with dilute nitric acid
	rinse with purified water
	dry with mouth downward
	Polyethylene bag for ash samples
	use clean bags (if in doubt, clean as above)
	heat seal (or self seal) in double pack (one bag inside another)
Shipment	air mail of international courier service
Accompanying data	letter for customs inspection*
	sample record sheets
	data sheets (analytical results, if available)

TABLE 14. GUIDELINES ON THE PREPARATION AND SHIPMENT OF SAMPLES TO THE CRL

\* Specimen letter

To whom it may concern

The enclosed samples are non-hazardous biological materials and are of no commercial value. They are for scientific research purposes only. They will be subjected to chemical analysis, during which they will be completely destroyed by ashing at a temperature exceeding  $400^{\circ}$ C.

(Signature block, i.e. name, position, organization, address, telephone, fax, e-mail).

For calculating z-scores, the relative standard deviation (RSD) of 20% was assumed for the results of the country laboratories and the CRL. The assumption of higher RSD was justified because of the known significant sample-to-sample biological variations in the concentrations of trace elements. As per the decision of the steering committee, wherever the z-scores were higher than 2.2, the country laboratory was advised to investigate into the possible reason for disagreement and repeat the analysis. In case the z-scores continued to be >2.2, the results were not considered for the final data evaluation. Z-scores are shown in Table 15 and as may be seen from the Table with the exception of Th, the z-scores for the other elements were less than 2. The element iodine was not included in z-score study since analysis of I for most samples obtained in Asian countries was not carried out by country laboratories but by a contract laboratory in Prague [44].

# TABLE 15: Z-SCORES FOR THE CUMULATIVE ANALYTICAL DATA FOR Cs, Sr, Th AND U ANALYSED AT THE COUNTRY LABORATORIES AND AT THE CRL (JAPAN)

	CHINA				INDIA	
Element	Mean $\pm$ ASD	Mean $\pm$ ASD	z-score	Mean ± ASD	Mean $\pm$ ASD	z-score
Cs (ng/g)	$47.3\pm9.4$	$65.4 \pm 13.1$	1.2	$11.0\pm2.2$	$10.8\pm2.2$	0.1
Sr (µg/g)	-	-	-	2.9 <u>+</u> 0.6	3.3 <u>+</u> 0.7	0.4
Th (ng/g)	$22.8\pm4.6$	$17.7 \pm 3.5$	0.9	$1.2\pm0.24$	$0.6 \pm 0.12$	1.6
U (ng/g)	9.4 ± 1.9	$11.4 \pm 2.3$	0.7	$0.8 \pm 0.2$	$1.6 \pm 0.3$	2.2
	PAKISTAN			I	REP. OF KOREA	
Cs (ng/g)	$12.1\pm2.4$	$12.3 \pm 2.5$	0.1	7.1 ± 1.4	$5.6 \pm 1.1$	0.8
Sr (µg/g)	$4.4\pm0.9$	$5.0 \pm 1.0$	0.4	$23.8\pm4.7$	$30.1 \pm 6.0$	0.8
Th (ng/g)	$6.2 \pm 1.2$	$6.8 \pm 1.4$	0.3	$4.9 \pm 1.0$	3.9 ± 0. 8	0.8
U (ng/g)	Uranium anal	lysis was carried ou	t at the CRI	$3.5 \pm 0.7$	$2.5\pm0.5$	1.2
	VIETNAM				JAPAN	
Cs (µg/g)	$21.9\pm4.4$	$28.4\pm5.6$	0.9	$98.0 \pm 1.0$	$98.0\pm3.0$	0.0
Sr (µg/g)	$5.8 \pm 1.2$	5.5 ± 1.1	0.2	$3.01 \pm 0.0$	$2.93\pm0.1$	0.6
Th (ng/g)	$12.8\pm2.6$	$6.3 \pm 1.3$	2.3	$8.28\pm0.01$	$8.0 \pm 0.5$	0.6
U (ng/g)	$1.3 \pm 0.3$	$3.7 \pm 0.7$	3.2	$2.3 \pm 0.4$	$2.2\pm1.4$	0.0

Analysis of only four elements of importance in radiation protection was carried out at country laboratories and at the CRL as part of the external QC.

In view of the extremely low concentrations of these elements in diet and tissue samples and the large variation in concentrations observed in the samples, a uniform Relative Standard Deviation (RSD) of 20% was assumed for the purpose of the z-score calculations.

For the CRL (Japan), the RSD was not assumed; instead the actual SD was taken for the calculation of the z-scores.

The data from Philippines and Bangladesh are not reported here as the analysis of the samples for the above reported elements was carried out at the CRL (Japan) only.

Indonesia could not complete the study on the above reported elements of radiological importance.

Z scores were not calculated for I, as its analysis for samples from all the countries was carried out under a separate technical contract (J. Kucera, Prague, Czech Republic).

### 7. RESULTS

With the exception of Bangladesh and Indonesia, all other Asian countries participating in the present CRP obtained dietary data on all five of the elements that had been defined as being of high priority. In Bangladesh dietary intakes of four and in Indonesia two of these five elements were studied. The elemental data obtained by various countries for essential and toxic elements, were also not similar. Analysis of total five elements in diet samples was in Indonesia and maximum of twenty-two elements were determined in diet samples in Vietnam. The details of elements studied and the number of diet samples analysed for each of these elements are shown in Table 16. Table 17 provides the information on 24 individual elements that were analysed in various Asian countries. The results on different types of samples collected and analysed for various elements in all the nine Asian countries are given below.

# TABLE 16. DETAILS OF ELEMENTS STUDIED AND NUMBERS OF SAMPLES ANALYSED IN DAILY DIETS BY THE PARTICIPATING COUNTRIES

Country	Important in Radiological Protection	Important in Nutrition	Toxic	Total
BANGALADESH	Ca(9) Cs (8) Sr(8) Th(9) U (8)	Cu(6) Fe(6) Mg(6) Mn(6) Zn(6)	-	10(72)
CHINA	Ca(48) Cs(48) I(48) K(48) Sr(48) Th(48) U(48)	Cu(48) Fe(48) Mg(48) Mn(48) Na(48) Zn(48) Se(48)	Cd(48) Hg(48) Pb(48)	17(816)
INDIA	Ca(20) Cs(20) I(10) K(20) Sr(20) Th(20) U(15)	Co(20) Cr(16) Fe(20) Se(17) Zn(20)	-	12(218)
INDONESIA	Ca(26) Cs(17) K(26) Sr(23)	Zn(19)	-	5(111)
JAPAN	Ca(27) Cs(27) I(54) K(27) Sr(27) Th(27) U(27)	Cu(27) Fe(27) Mg(27) Mn(27) Na(27) P(27) Zn(27)	-	14(405)
REPUBLIC OF KOREA	Ca(5) Cs(5) I(4) K(5) Sr(5) Th(5) U(5)	Zn(5)		8(39)
PAKISTAN	Ca(31) Cs(31) I(31) K(31) Sr(31) Th(22) U(12)	Cr(28) Fe(31) Mg(31) Mn(8) Na(31) Se(30) Zn(31)	Hg(26) Sb(20)	16(425)
PHILLIPPINES	Ca(19) Cs(31) I(19) K(19) Sr(31) Th(31) U(31)	Cr(19) Fe(19) Mg(19) Mn(19) Na(19) Se(19) Zn(19)	-	14(314)
VIETNAM	Ca(18) Cs(18) I(18) K(18) Sr(18) Th(18) U(18)	Ba(18) Co(18) Cu(18) Fe(18) Mg(18) Mn(18) Na(18) Ni(18) P(18) Zn(18)	As(18) Cd(18) Hg(18) Pb(18)	22(396)
TOTAL — ALL COUNTRIES				2796

S. No.	Element Country T										Total
Radio- logical	Element	Bangla- desh	China	India	Indo- nesia	Japan	Pakistan	Philip- pines	Republic of Korea	Viet nam	
1	Са	+	+	+	+	+	+	+	+	+	9
2	Cs	+	+	+	+	+	+	+	+	+	9
3	Ι	-	+	+	-	+	+	+	+	+	7
4	K	-	+	+	+	+	+	+	+	+	8
5	Sr	+	+	+	+	+	+	+	+	+	9
6	Th	+	+	+	-	+	+	+	+	+	8
7	U	+	+	+	-	+	+	+	+	+	8
Minor											
8	Mg	+	+	-	-	+	+	+	-	+	6
9	Na	-	+	-	-	+	+	+	-	+	5
10	Р	-	-	-	-	+	-	+	-	+	3
Trace- essential											
11	Со	-	-	+	-	-	-	-	-	+	2
12	Cr	-	-	+	-	-	+	-	-	+	3
13	Cu	+	+	-	-	+	-	+	-	+	5
14	Fe	+	+	+	-	+	+	+	-	+	7
15	Mn	+	+	-	-	+	+	+	-	+	6
16	Se	-	+	+	-	-	+	-	-	-	3
17	Zn	+	+	+	+	+	+	+	+	+	9
Toxic											
18	As	-	-	-	-	-	-	-	-	+	1
19	Cd	-	+	-	-	-	-	-	-	+	2
20	Hg	-	+	-	-	-	+	-	-	+	3
21	Pb	-	+	-	-	-	-	-	-	+	2
22	Sb	-	-	-	-	-	+	-	-	-	1
Others											
23	Ba	-	-	-	-	-	-	-	-	+	1
24	Ni	-	-	-		-	-	-	-	+	1
Total	24	10	17	12	5	14	16	14	8	22	

# TABLE 17. ELEMENTS DETERMINED IN DIET SAMPLES IN EACH ASIAN COUNTRY

#### 7.1. Bangladesh

#### 7.1.1. Ingestion study

In Bangladesh, a countrywide statistical dietary survey was carried out on the food consumption. This formed part of the nutritional survey of rural and urban areas of Bangladesh [45] covering 14 locations in the country and about 50 families at each location, who belonged to different income groups. In all, 4315 subjects in the age group of 20–50 years were studied and eleven main food components consumed by the population were identified [46]. On the basis of the average consumption of these 11 food components in diet, raw food materials were purchased from the markets in and around Dhaka.

The diets were prepared from these food materials in a cooking style typical of Bangladesh. These diet samples were homogenized using mixer blenders. The diet samples in Bangladesh were not freeze dried but oven dried at 100–110°C instead. Therefore iodine, which is volatile in nature and could escape from samples during oven drying, was not studied.

In Bangladesh, the diet samples collected for the study were analysed for ten elements. Out of them Ca, Cu, Fe, Mn, Mg and Zn were analysed within the country itself using AAS, and for Cs, Sr, Th and U the aliquots of the samples were sent to the CRL for analysis using ICP-MS and ICP-AES.

#### 7.1.2. Daily dietary intakes

In all, 72 determinations were made for ten elements in 6–9 diet samples. Based on their concentrations daily dietary intakes were estimated. The results are shown in Table 18. Intra-country variations for dietary intakes of Th and U were more than an order in magnitude. For the other elements studied, the variation was minimum (1.5 times) for Ca and maximum (up to 4 times) for Cs.

TABLE 18. DAILY DIETARY INTAKES OF TRACE ELEMENTS BY THE ADULT POPULATION OF BANGLADESH

Element	No. of samples	Range	Mean	S.D.	Median
Ca (g)	9	0.25-0.35	0.29	0.05	0.31
Cs (µg)	8	11.01-40.75	24.09	11.42	23.58
Sr (mg)	8	1.02-2.56	1.79	0.45	1.83
Th (µg)	9	1.33–16.99	5.03	5.16.	3.52
U (µg)	8	0.51-8.18	2.33	2.56	1.31
Cu (mg)	6	0.68–2.23	1.80	0.57	1.95
Fe (mg)	6	8.30-18.02	12.10	3.50	10.80
Mg (g)	6	0.21-0.48	0.32	0.10	0.32
Mn (mg)	6	2.01 - 4.08	3.23	0.84	3.38
Zn (mg)	6	3.92-12.37	8.70	2.72	9.07

#### 7.1.3. Organ content study

Bangladesh is predominantly a Muslim country and because of the society norms prevailing there, the tissue samples were not collected for the organ content study.

#### 7.2. China

#### 7.2.1. Ingestion study

Diet samples in China were collected according to the sampling strategy devised for the 'First Total Diet Study in China' in 1990 by the Institute of Nutrition and Food Hygiene, Chinese Academy of Preventive Medicine [47]. Based on geographic location, dietary habits and cooking style, the whole of China was divided into 4 regions and from each region one province was selected for the survey. In

each of these provinces, 3 survey points (one town and two countryside) were identified, from where 30 families were randomly chosen. The diet composition and the consumption of various food items were recorded by means of weight record for three successive days. Some of these details are given in second total diet study, carried out in China in 1992 [48]. The total diet composition as well as the average daily consumption of each food for Chinese adults, engaged in light physical activity was calculated for each region. Based on the composition of diet and individual food materials, all foods and the products derived from them were classified into the following 13 types: (1) Grain; (2). Beans and nuts; (3) Yam; (4) Meat (including poultry); (5) Egg; (6) Aquatic foods; (7) Milk; (8) Vegetable; (9) Fruits and its salads, etc.; (10) Sugar; (11) Soft beverages and drinking water; (12) Alcoholic beverages; (13) Spices and cooking oil. These foods were collected in 1997 from nearby vegetable markets, subsidiary food stores and farmer markets within the three-survey points in each region. Various foods thus collected were treated and cooked according to the local dietary habit in assigned restaurants and kitchens. Cooked foods were then ground and mixed in the blender. In all, 48 individual food samples were obtained representing the four regions. Samples under frozen condition were shipped to the analytical laboratory for further processing and analysis. All possible precautions already stated in the sampling and quality control chapter were taken to avoid contamination of samples.

Analysis of diet samples was carried out using ICP-MS for Cs, Th and U, ICP-AES for Ca, Cu, Mg, Mn, Sr, INAA for Fe and Zn, ENAA for I, AAS for K, Na and Cd, fluoro-photometry for Se, and cold-vapor-AAS for Hg.

# 7.2.2. Daily dietary intake in China

The results of the intake study are given in Table 19. Concentrations of 17 elements (of importance in radiological protection, nutrition, and a few toxic elements Cd, Hg and Pb) were determined in diet. Daily intakes of Ca, Cs, I, K, Sr, Cu, Mg, Se, Zn, Pb showed variation by a factor of <2 and that for the elements Fe, Na and Cd was between 2–3. The variation in intake of toxic element Hg was up to a factor of 5. The results of three toxic elements Pb, Cd and Hg although included in the table, will not form part of the discussion.

# 7.2.3. Contribution of individual food materials to daily intake

The individual food materials, which contribute to the daily diet were also analysed to obtain information about the percentage contribution of these foods materials to the daily intakes of trace elements. The results shown in Tables 20 and 21 respectively reveal that the major contribution to intakes of Ca, Cs, I, K, Sr, came from cereals and vegetables. For Th however, the main contributors were grains followed by vegetables and then by beans. For U, cereals and then drinking water contributed significantly to the daily intake. Grains were the main contributors of: Fe, Zn and Mn.

# 7.2.4. Organ content study

By multiplying average weights of relevant organs in Chinese reference man and tissue concentrations of trace elements, organ contents were estimated. The content of I in thyroid, Cs in skeletal muscle, Sr in skeleton and Th and U, in liver, kidney, lung and skeleton were estimated for the Chinese population. The results are shown in Table 22. With the exception of content of Ca in skeleton and I in thyroid, the variation in the organ content of most of the elements was one to two orders of magnitude.

Organ contents of a large number of some other trace elements (not included in the scope of the project) were also determined for the Chinese population. But only the data related to the elements of interest in radiological protection have been included in this report.

Element	No. of samples*	Range	Mean	S.D.	Median
Ca (g)	48	0.62-0.91	0.72	0.13	0.68
Cs (µg)	48	10.33-15.51	12.95	2.12	12.98
I (µg)	48	175.2–631.9	365.2	198.90	326.8
K (g)	48	1.69–2.56	2.11	0.43	2.09
Sr (mg)	48	1.99–3.67	2.86	0.71	2.88
Th (µg)	48	1.14-14.07	5.37	5.88	3.13
U (µg)	48	1.56–15.36	6.75	6.43	5.04
Cu (mg)	48	1.29–2.31	1.60	0.48	1.39
Fe (mg)	48	16.90-44.90	30.50	13.60	30.10
Mg (g)	48	0.26-0.45	0.36	0.08	0.36
Mn (mg)	48	5.36-8.58-	6.82	1.333	6.67
Na (g)	48	1.96–5.11	3.64	1.33	3.75
Se (µg)	48	43.1-63.8	52.8	9.7	52.0
Zn (mg)	48	11.66–14.22	12.45	1.21	12.01
Cd (µg)	48	37.90–106.20	75.60	29.60	79.0
Hg(µg)	48	12.60–59.30	34.50	19.30	33.00
Pb (µg)	48	286.0-563.0	391.0	133.0	357.0

TABLE 19. DAILY DIETARY INTAKES OF TRACE ELEMENTS BY THE ADULT POPULATION OF CHINA

\*12 Food types from 4 regions and each region had 3 survey points where 30 families were randomly chosen.

# TABLE 20. PERCENTAGE CONTRIBUTION OF DIFFERENT FOODS TO DIETARY INTAKES OF TRACE ELEMENTS IN CHINA

Food Item	Ca	Cs	Ι	К	Sr	Th	U
Grains	39.6	40.8	23.3	35.9	39.2	69.8	55.1
Beans	9.3	9.4	6.3	5.5	7.7	4.5	3.7
Yam	1.6	5.6	9.7	6.7	2.4	2.6	2.8
Meat	1.1	9.9	7.0	5.1	2.1	2.0	1.5
Egg	1.4	0.7	2.1	0.8	0.7	0.0	0.1
Aquatic products	8.0	2.3	4.5	3.7	2.8	1.3	1.0
Milk	3.3	0.5	0.3	0.9	0.0	0.0	0.0
Vegetables	31.0	25.9	43.4	38.7	38.1	17.1	11.0
Fruit	0.4	1.2	0.1	1.9	0.7	0.2	0.1
Sugar	0.0	0.2	0.0	0.0	0.0	0.0	0.0
Drink & water	4.3	3.4	3.2	0.5	6.3	2.0	24.4
Alcoholic drinks	0.0	0.1	0.0	0.0	0.0	0.0	0.1

Item	Cu	Fe	Mg	Mn	Na	Se	Zn
Grains	66.9	59.0	55.0	71.8	15.1	61.4	66.6
Beans	15.6	6.9	9.6	8.1	4.4	1.9	5.8
Yam	1.9	3.3	2.0	1.3	6.0	0.9	1.1
Meat	1.2	4.6	2.8	0.6	8.7	7.8	8.4
Egg	0.6	1.0	0.6	0.0	1.8	4.5	1.4
Aquatic products	0.6	2.6	3.4	1.5	4.0	15.5	3.4
Milk	0.0	0.0	0.6	0.0	0.1	0.6	0.5
Vegetables	10.6	20.6	20.3	13.0	59.6	6.1	11.1
Fruit	1.2	0.3	0.8	1.0	0.0	0.0	0.2
Sugar	0.0	01.3	0.0	0.0	0.0	0.0	0.0
Drink & water	0.6	0.3	4.5	2.6	0.4	0.9	1.4
Alcoholic drink	0.0	0.0	0.3	0.0	0.0	0.0	0.0

# TABLE 21. PERCENTAGE CONTRIBUTION OF DIFFERENT FOODS TO DIETARY INTAKES OF TRACE ELEMENTS IN CHINA

# TABLE 22. CONTENTS OF SOME SELECTED ELEMENTS IN MAIN TISSUES AND ORGANS OF MALE CHINESE ADULTS

Organ	No. of samples	Element	Range	Mean	S.D.	Median
Skeleton	30	Ca (g)	566-1520	987	964	976
Skeletal Muscle	31	Cs (µg)	134–1870	552	367	469
Thyroid	31	I (mg)	6.61–36.9	18.6	6.61	19.3
Skeleton	30	Sr (mg)	6.4–1069	457	273	353
Lungs	31	Th (µg)	0.34–50.2	11.9	12.0	7.8
Liver	31	Th (µg)	0.11–6.78	0.59	1.42	0.53
Skeleton	31	Th (µg)	2.22–138	20.9	24.3	14.4
Kidney	31	U (µg)	0.01–1.10	0.29	0.24	0.26
Lungs	31	U (µg)	0.29–10.2	2.84	2.59	1.92
Liver	31	U (µg)	0.25–7.27	1.60	1.70	0.77
Skeleton	31	U (μg)	4.67–70.7	19.5	16.2	12.6

# 7.3. India

The reference Asian Man Phase-2 study in India included investigations of the daily dietary intakes of fourteen elements of importance in radiological protection and nutrition, analysis of individual food materials and estimation of the organ contents for elements Cs, I Sr, Th and U in relevant source organs (where these elements and related radionuclides concentrate).

# 7.3.1. Ingestion study

Indian population is large and heterogeneous and therefore cannot be represented by 20–30 duplicate diet samples. Extensive surveys conducted by the National Nutrition Monitoring Board (NNMB) of India [49–51] were utilised to obtain representative intake data on various food items. Dang et al. [52], have used data provided by NNMB for the rural and urban population of the country, to identify and propose various food materials (groups) and their quantities consumed by the adult Indian. The perishable food materials in appropriate quantities were purchased from local market whereas the cereals and pulses were purchased for four provinces located in four zones of the country based on the procedure established earlier by Dang et al. [53]. The diet samples were then prepared in the typical cooking style prevailing in the country. The aliquots of diet samples were then homogenized, freeze dried, powdered and taken up for analysis.

# 7.3.2. Daily dietary intake in India

The analysis of samples for Cs, Ca, K, Fe, Co, Se, Cr, and Zn was carried out using INAA. Th, U and Sr were determined by RNAA and I by using both RNAA and ENAA From data on concentrations of trace elements in total diet samples as well as in food materials, the daily intakes and the contributions from the food ingredients to the daily intakes of various trace elements were estimated. The results on the daily intakes of 12 trace elements by reference Indian Adult male are given in Table 23. As may be seen from the Table, the intra-country variations in the intake of twelve trace elements ranged between 3 to 5.

Element	No. of samples	Range -	Daily Intake		
Element	No. of samples	Kange	Mean	S.D.	Median
Ca (g)	20	0.17–0.67	0.37	0.14	0.36
Cs (µg)	20	2.64-11.8	5.22	2.61	4.35
Ι (μg)	10	56.90-260.0	123.30 (+120) (salt)	67.40	85.00 (+120)
K (g)	20	1.24-3.84	1.98	0.84	1.57
Sr (mg)	20	0.76-2.96	1.59	0.68	1.50
Th (µg)	20	0.44-1.75	0.82	0.39	0.64
U (µg)	15	0.35-1.16	0.58	0.28	0.54
Co (µg)	20	8.3–31.4	17.9	7.48	16.99
Cr (µg)	16	27.8-105.4	59.9	22.3	54.90
Fe (mg)	20	10.2-34.4	17.04	6.92	14.85
Se (µg)	17	35.0-130.6	57.40	29.30	43.50
Zn (mg)	20	5.3-15.60	9.12	3.46	7.86

TABLE 23. DAILY DIETARY INTAKES OF TRACE ELEMENTS BY ADULT INDIAN POPULATION

(+120) is the possible contribution made by the iodized salt used for cooking purposes.

#### 7.3.3. Contribution of individual food materials to the daily intake

About 3–7 samples of the individual food ingredients (in duplicate or triplicate) were analysed for trace elements stated above. Using data on concentrations and quantities of the individual food materials consumed, their contribution to the daily intakes were calculated. The results are shown in Table 24.

It was observed that for Th, U, Fe, Zn, Co, Se, Cr etc., the maximum contribution to their daily intakes came from the consumption of cereals and pulses (legumes). The Cs intake was contributed equally by cereals, pulses and milk. It was interesting to note from the results that due to higher concentration of Cs in pulses (legumes), despite their consumption in small quantity by the adult population of India, the contribution of Cs from it was quite considerable. Iodine mainly came from iodized salt, and intake of Ca was contributed by milk and green vegetables. Some other food materials such as fruits, spices, jaggery etc. were not analysed and their contributions to the daily intake are included in the category of other foods.

TABLE 24. TYPICAL DAILY INTAKES OF TRACE ELEMENTS FROM IMPORTANT FOOD COMPONENTS OF THE DIET OF THE ADULT INDIAN POPULATION

Element	Rice	Wheat	Sorghum Vulgare	Total cereals	Legumes	Milk	Tuber/ Roots	Green Veg.	Others	Total	Other Foods
Ca (mg)	(.54 (2.7)	26.4 (7.5)	6.0 (1.7)	41.94 (11.9)	14.4 (4.1)	9.3 (19.8)	5.9 (1.7)	60.0 (18.9)	8.3 (2.4)	80.2 (23.0)	51.2
Cs (µg)	0.69 (14.5)	0.19 (4.0)	0.51 (10.7)	1.39 (29.2)	0.97 (20.4)	0.52 (10.9)	).06 (1.3)	0.15 (3.2)	).05 (1.0)	0.26 (5.3)	34.2
I (µg)	21.8 (9.5)	6.6 (2.9)	-	-	-	8.64 (3.8)	-	-	-	-	
K (mg)	278.2 (15.1)	203.0 (11.0)	299.0 (16.2)	780.0 (42.3)	217.6 (11.8)	*1.0 (4.4)	1117.0 (6.4)	65.0 (3.5)	58.9 (3.2)	240.8 (13.1)	28.4
Sr (µg)	37.1 (2.5)	225.0 (15.4)	375.0 (25.7)	637.1 (43.6)	83.2 (5.7)	53.0 (3.6)	37.2 (1.0)	4.5 (0.3)	43.5 (3.0)	85.2 (4.3)	42.4
Th (ng)	214.6 (28.7)	85.2 (11.4)	92.00 (12.3)	391.8 (52.4)	30.7 (4.1)	4.5 (0.6)	10.75 (1.4)	30.0 (4.0)	10.9 (1.4)	51.7 (6.8)	36.1
U (ng)	74.2 (12.7)	57.6 (9.8)	74.0 (12.6)	205.8 (35.1)	32.3 (5.5)	21.9 (3.7)	6.45 (1.1)	20.8 (3.5)	7.0 (1.2)	33.8 (5.8)	49.9
Co (µg)	5.5 (32.3)	1.1 (6,5)	0.2 1.1)	6.8 (40.0)	0.7 (4.1)	0.1 (0.6)	0.41 (2.4)	0.23 (1.4)	0.23 (1.4)	1.54 (9.1)	46.2
Cr µg)	-	23.4 (41.7)	-	-	-	-	-	-	-	-	
Fe (mg)	1.25 (7.9)	2.5 (15.7)	1.8 (11.3)	5.5 (34.9)	1,25 (7.9)	0.1 (0.6)	0.18 (1.1)	0.75 (4.7)	0.13 (0.8)	1.06 (6.6)	50.0
Se (µg)	7.8 (14.9)	3.9 (7.4)	-	-	1.6 (3.0)	-	-	-	-	-	
Zn mg)	2.7 (31.5)	1.4 (16.3)	1.1 (12.8)	5.2 (60.7)	0.91 (10.6)	0.4 (4.7)	0.06 (0.7)	).15 (1.7)	0.09 (1.0)	0.30 (3.5)	20.5

The number in the parentheses denotes the percentage contribution of the individual foods.

#### 7.3.4. Organ content study

Organ contents of Cs, I, Sr, Th and U were obtained from the concentrations of these elements in tissue materials and the organ weight data for reference Indian Man obtained for the RAM Phase I of the CRP. The human tissue samples collected for the organ content study were thyroid for I, lung and liver for Th and U, skeletal muscle for Cs, kidney for U and Th and bone for Th, U and Sr. From 10-20 human tissue samples. Up to 3 aliquots were analysed to obtain the concentrations of the trace elements in them. The results are shown in Table 25. The organ content of the five elements in adult Indian population varied by a factor ranging from 3 for iodine in thyroid to more than 15 for U in bone. The minimum variation was for the Th content of liver (<3).

Organ	No. of samples	Element	Range	Mean	S.D.	Median
Thyroid (mg)	14	Ι	4.36–12.6	6.97	2.26	6.27
Kidney (µg)	15	U	0.08–0.48	0.21	0.09	0.19
Lungs (µg)	15	U	0.50–2.64	1.43	0.80	1.02
Liver (µg)	21	U	0.03–0.15	0.08	0.04	0.08
Skeleton (µg)	15	U	0.65–12.1	4.20	3.70	2.85
Lungs (µg)	23	Th	1.20-9.68	5.18	3.06	4.35
Liver (µg)	9	Th	0.09–0.23	0.15	0.06	0.12
Skeleton (µg))	19	Th	1.29–12.9	4.11	2.90	3.96
Skeleton (mg)	11	Sr	42.5–580	235.7	150.0	172.5
Skeletal Muscle (µg)	10	Cs	153.–670	330.0	144	327.0
Skeletal Muscle (g)	18	К	18.7–87.4	54.4	21.5	54.9

# TABLE 25. CONTENTS OF SOME SELECTED ELEMENTS IN MAIN TISSUES AND ORGANS OF ADULT INDIAN POPULATION

#### 7.4. Indonesia

#### 7.4.1. Ingestion study

The study in Indonesia was carried out on a very small scale both for ingestion and organ contents of elements. The intakes of Cs, Sr, Ca and K in the group of elements of radiological importance and only Zn among essential elements were estimated. As part of the organ contents study, only 3 bone samples were analysed for K, Ca and Sr. In view of the very small number of samples for one individual organ, the results were not considered for inclusion in the main document.

The food samples were prepared on the basis of the daily per capita consumption of calorie data. These data for food commodities were published in Indonesia by Central Bureau of Statistics [54] and the energy values per gram of individual vegetables, fruits, meat etc, were published by Nutrition

Research and Development Centre in Indonesia [55]. Some duplicate diet samples were also collected for the study. These duplicate samples were generally collected from canteens and eating houses at the inter-city bus stops. Although duplicate diets were processed and analysed as complete one day samples, for the market basket study on the other hand, only one tenth of the food material was used to prepare the food for analysis. The samples from Indonesia were not collected as per the IAEA protocols, which stated the preparation of total diet representative of one day's meal of an adult. Further, the duplicate diet samples were obtained from canteens and therefore did not represent the exact duplicate portion of diet of the individual subjects representing the adult population of Indonesia.

These food samples were blended, freeze dried and subsequently powdered with glass mortar prior to analysis. AAS and INAA were employed to carry out the analysis of diet samples for various trace elements.

# 7.4.2. Daily dietary intake in Indonesia

In Indonesia, the data on daily intakes of only Sr, K, Ca, Cs and Zn were collected. 17–26 samples of diet were collected, processed and analysed for these five elements. Results of daily dietary intakes in Indonesia are shown in Table 26. Large variations in daily intakes of Cs, K and Sr (more than an order in magnitude) were observed. The variations in the intakes of Ca and Zn were relatively smaller. This may be due to non-adherence of quality control proposed by the IAEA and followed in other Asian countries. The data however provided useful information and therefore have been included under this section on results.

# TABLE 26. ESTIMATED DAILY INTAKES OF K, Ca, Sr, Cs AND Zn IN THE DIET OF ADULT POPULATION OF INDONESIA

Element	No. of samples	Range	Mean	SD	Median
K (g)	26	0.2–2.8	1.34	1.32	1.1
Ca (g)	26	0.1 - 1.0	0.3	0.25	0.3
Sr (mg)	23	0.2–7.5	2.4	1.7	2.2
Zn (mg)	19	0.8–5.9	2.95	1.49	2.5
Cs (µg)	17	1.6-77.0	27.8	22.9	15.4

# 7.4.3. Organ content study

Indonesia is also a predominantly Muslim country and issues in this context discussed earlier affected the collection of tissue sample. Only 3–4 skeletal tissue samples were collected processed and analysed for Ca, Sr and K. The results are shown in Table 27. Since the number of samples was small, no comments are offered on the variations observed for these three elements.

# TABLE 27. THE CONTENTS OF THREE IMPORTANT TRACE ELEMENTS IN SKELETON OF ADULT INDONESIAN MALE

Element	Elemental Content of skeleton							
	No. of samples	Range	Mean	S.D.	Median			
K (g)	4	7.65–22.1	14.9	5.95	14.5			
Ca (g)	3	722.5–1436.5	971.6	47.4	940.0			
Sr (g)	3	0.85–0.85	0.85	0	0.85			

# 7.5. Japan

Japan is a developed country in the Asian region with very little or no population below the poverty line. The food intake pattern of the Japanese population is believed to be quite uniform and hence it was possible to use duplicate diets to study dietary intakes of trace elements by country's adult population. Duplicate diet samples were collected from nine different regions spread over the entire country considering the comparatively homogeneous socio-economic status of the Japanese population. These samples covered diets consumed on three consecutive days from nine subjects in the age range of 20–50 a. In addition, food materials were also collected to develop the Typical Japanese Diet RM (see section 6 on QA/QC).

# 7.5.1. Ingestion study

Twenty-seven duplicate diet samples were analysed to estimate the daily intakes of 13 elements. The only exception was iodine, for which 54 samples were analysed. The samples after collection were homogenised using a blender fitted with Titanium coated blade. The elements determined were Ca, Cu, Fe, I, K, Mg, Mn, Na, P, Zn, Sr, Cs, Th and U. The analysis of the diet for 14 elements was carried out mainly by ICP-MS and ICP-AES. The only exception was I which was analysed by using ICP-MS, INAA and ENAA.

#### 7.5.2. Daily dietary intake

In all, 405 determinations were made for 14 elements on these 27 diet samples. Results for the range, mean (SD) and median values of dietary intake by the Japanese reference man are given in Table 28. For some of the samples, the iodine intake was observed to be quite high. The relative standard deviation was up to 200% and the mean value was four times larger than the median value. Therefore the median is likely to be a better representative of the intake of I in Japan instead of the mean value as the median does not get significantly affected by the extreme values. Except for the minor elements Na, K and P, the intra-country variations for other elements are quite significant. The intake of Fe, and to some extent the intake of Cu, was observed to be quite low for the Japanese adult population when compared to the intakes in Pakistan or China.

#### 7.6. Pakistan

#### 7.6.1. Ingestion study

In Pakistan, estimation of daily dietary intakes of 16 trace elements by the adult population was carried out. These included elements of both radiological and nutritional importance as well as some toxic elements such as Sb, Hg etc. The data are presented in Table 29.

Diet samples were prepared on the basis of the market basket study. The mean dietary consumption of individual food components by Pakistan reference man was used to prepare the diet. A slight modification in the preparation of the meal was adopted. All the food components in the amount eaten by the representative population of Pakistan were mixed and then cooked for a few minutes in a microwave oven. 250 mg aliquots of the dried powdered food samples were drawn for elemental analysis.

Element	No. of samples	Range	Mean	S.D.	Median
Ca (g)	27	0.12-1.29	0.50	0.27	0.44
K (g)	27	1.15–2.62	1.91	0.41	1.86
Cs (µg)	27	3.51-19.51	7.91	3.24	7.01
Sr (mg)	27	0.52-3.86	1.68	0.84	1.60
I (μg)	54 *	38-8710	1020	1964	246.0
Th (µg)	27	0.12-0.97	0.35	0.20	0.32
U (µg)	27	0.13-6.52	1.10	1.42	0.64
P (g)	27	0.52-1.26	0.82	0.18	0.79
Mg (g)	27	0.12-0.27	0.20	0.04	0.21
Na (g)	27	2.07-5.27	3.98	0.65	4.13
Fe (mg)	27	2.80-15.87	5.43	2.44	4.91
Cu (mg)	27	0.44–1.73	0.99	0.34	1.06
Mn (mg)	27	1.34–5.45	3.44	0.94	3.51
Zn (mg)	27	4.66-12.75	8.35	2.29	8.27

TABLE 28. DIETARY INTAKES OF 14 ELEMENTS BY JAPANESE ADULT POPULATION

\* Including 27 samples analysed by ICP-MS (Muramatsu, 2003).

TABLE 29: DAILY DIETARY INTAKES OF SOME IMPORTANT TRACE ELEMENTS BY THE ADULT POPULATION OF PAKISTAN

Elements	No. of samples	Range	Mean	SD	Median
Sr (mg)	31	0.91-5.70	2.93	1.44	2.43
Ca (g)	31	0.22-0.71	0.51	0.14	0.48
K (g)	31	1.7-4.3	2.69	0.54	2.62
Cs (µg)	30	2.6-12.9	6.16	2.59	5.97
Th (µg)	22	0.92-7.15	3.01	1.67	2.66
U (µg)	12	1.35-6.70	3.06	1.80	2.16
Mg (g)	31	0.22-0.92	0.50	0.15	0.52
Na (g)	31	3.32-3.65	3.43	0.07	3.42
Fe (mg)	31	12.46-51.61	31.04	8.97	30.03
Zn (mg)	31	7.62-23.12	13.59	3.31	14.25
Sb (µg)	20	0.42-11.48	1.64	2.40	1.00
Cr (µg)	28	42.8-370.4	129.9	71.4	118.90
Hg (µg)	26	0.78-6.5	2.59	1.47	2.36
Se (µg)	30	30.6-270.2	108.5	55.4	103.5
Mn (mg)	8	3.7-24.36	10.57	6.75	11.75
I (µg)	31	15.4-139.2	47.34	29.47	43.47

# 7.6.2. Daily dietary intake

A number of different analytical techniques, namely INAA, AAS, ICP-AES and ICP-MS, were employed. Na, K, Mg and Fe were analysed using AAS, Sr and Mn by ICP-AES and Ca was analysed using both AAS and ICP-AES. The dietary intakes of the elements Th, Zn, Se, Sb, Hg, Cs, Cr were estimated using INAA. U was analysed using ICP-MS and I was analysed by ENAA/RNAA in a contract laboratory arranged by the IAEA. Quality control was carried out as described in the earlier Sections.

Diet samples ranging in number from 22–31 were analysed for most elements with the exception of a few elements like U(12), Mn(8), Sb(20), where a smaller number of samples (number given in parentheses) were analysed. The results are shown in Table 29. The salient feature of the intakes study is the markedly low intake of iodine and the high intake of Fe by the population of Pakistan. The variations in the intakes of most elements are not large except for I, Se, Cr and Sb, for which the intakes by different sections of the population vary by an order of magnitude.

#### 7.6.3. Organ content study

The study on organ contents of trace elements could not be carried out, due to cultural norms prevalent in that country, which do not allow autopsy.

#### 7.7. Philippines

In addition to the dietary intake of trace elements of radiological and nutritional importance, ten samples each of human liver and lung tissues were also analysed for Th, U and Cs for the adult subjects from the Philippines. The study of the elements of importance in nutrition assumed greater significance in the Philippines in view of the reported malnutrition in all the 13 regions in the country, including the National Capital Region. The Food and Nutrition Research Institute (FNRI), the agency responsible for monitoring the nutrition status of the Philippines, is currently refocusing its studies on the mineral and elemental content of food.

# 7.7.1. Ingestion study

One-day diet samples per region were prepared from the commonly eaten foods as documented by the FNRI [56]. Only the edible portions of raw foods were processed to obtain food samples and the diets were prepared using regional recipes of A. Mondala [57]. The test meal consisted of breakfast, lunch, dinner and one snack. Selection was based on the low cost, availability of food items and ease of preparation. The food also included food items eaten raw and also as processed food and beverages. The meal was cooked from edible portions of raw food materials purchased from market. The other types of sampling was the purchase of (regional) cooked meals in the quantity and selection recommended by FNRI. The dry weights of the diet samples ranged from 130–401g (freeze dried weights), with a mean dry weight of 300 g.

The analysis of naturally occurring radionuclides Th and U along with the trace elements Sr and Cs was carried out using ICP-MS and elements P, Ca, Fe, Mn, Mg, Cu, Zn along with the electrolytes Na and K were all analysed using ICP-AES. The analysis of all samples from Philippines for all elements except I, was carried out at NIRS, Japan. I was determined using ICP-MS at NIRS and also by using ENAA in a contract laboratory at Prague arranged by the IAEA.

### 7.7.2. Daily dietary intake

The results of dietary intake of 14 elements are shown in Table 30. There were very large intracountry variations of intakes of different elements: more than 40 times for U, 50 times for Th, 25 times for I, and more than 10 times for Cs. Even the essential elements such as Cu, Fe and Mn showed fairly large variations in dietary intakes. The variations in intakes for Ca, Mg, Na, Zn, however, were relatively small. No analytical quality control was carried out in the country laboratory since analysis of all the samples for all the elements was carried out at NIRS Japan. Therefore, the QC aspects may be considered similar to the results generated by the CRL.

#### 7.7.3. Organ content study

Ten samples each of liver and lung tissues were obtained from accidental deaths during the last quarter of 1999. These twenty autopsy samples belonged to adult male Filipinos, whose ages ranged from 24–45 years and classified as normal. The tissue samples collected, weighed from 150 to 250 g in fresh weight. These tissue samples were sliced into thin pieces, packed, weighed and were freeze dried. The contents of various elements in ten samples of liver and lung of the Filipino adult population are given in Table 31. It is seen that the contents of Th and U in liver and lungs of the Philippine population lie in the range of organ content values obtained from the other Asian countries.

Trace Elements	No. of Samples	Range	Mean	S.D.	Median
Ca (g)	19	0.11-0.69	0.30	0.13	0.22
Cs (µg)	31	2.56-28.13	8.30	6.75	5.25
I (µg)	19	21.5-520	121.0	120.65	40.0
K (g)	19	0.55-1.81	1.04	0.33	1.0
Sr (mg)	31	0.69–5.15	1.57	0.83	1.36
Th (µg)	31	0.02–1.69	0.41	0.50	0.15
U (µg)	31	0.11-4.82	0.4	1.06	0.56
Cu (mg)	19	0.66–3.71	1.19	0.68	0.95
Fe (mg)	19	2.44-43.84	8.35	9.8	4.22
Mg (g)	19	0.06-0.21	0.14	0.04	0.13
Mn (mg)	19	0.95–6.76	2.83	1.54	2.34
Na (g)	19	1.22–2.78	1.96	1.93	1.14
P (g)	19	0.30–0.84	0.56	0.14	0.56
Zn (mg)	19	2.35-8.17	5.16	1.73	5.23

TABLE	30.	DAILY	DIETARY	INTAKES	OF	IMPORTANT	TRACE	ELEMENTS	BY	THE
ADULT	POP	ULATIO	N OF THE	PHILIPPINE	ES					

Element and Units	Organ	Content	Content of various trace elements in human organs					
	organ	No. of samples	Range	Mean (SD)	Median			
Cs (µg)	Liver	10	21.7-54.7	34.3	32.1			
Cs (µg)	Lungs	8	8.3–19.1	13.7 (3.7)	13.5			
Th (µg)	Liver	10	0.09–0.36	0.20 (0.10)	0.24			
Th (µg)	Lungs	8	0.43-2.10	0.87 (0.7)	0.64			
U (µg)	Liver	10	0.04-0.21	0.10 (0.05)	0.12			
U (µg)	Lungs	8	0.21–0.67	0.36 (0.2)	0.36			

# TABLE 31. ORGAN CONTENTS OF SOME IMPORTANT TRACE ELEMENTS IN TISSUES OF THE ADULT PHILIPPINE POPULATION

# 7.8. Republic of Korea

#### 7.8.1. Ingestion study

The samples of one day total diet of an adult Korean, based on the food intake data survey of 108 healthy subjects in the age range 20–50 a were prepared in typical Korean style for the intake study. The sampling was based on market basket concept. In all, five samples of total cooked diet were prepared. The samples were thoroughly mixed in a mixer blender fitted with Titanium coated blade and 20% of it was drawn from the lot for freeze drying. The analytical methods used for Cs, U, Th, Sr, I were a combination of INAA, RNAA as well as ICP-MS. For the determination of Ca, Cs, K, Sr and Zn, INAA/ICP-MS were employed and the rest of elements were analysed using both RNAA and ICP-MS.

Four most commonly consumed foodstuffs in the Republic of Korea, namely Kimchi, fish, pork and beef combination were also analysed to determine their contribution to the daily elemental intakes. However, the four commonly consumed foodstuffs from Korea did not represent the total intake in terms of the major or micronutrients. The data inputs from these food materials were therefore not considered for the Asian diet. They however represent important sources of trace element intakes in Korean diet and this information will be included in another technical document to be prepared at the IAEA with compilation of final country reports on the CRP from all the participating countries.

# 7.8.2. Dietary intake study

The daily dietary intake of eight elements Ca, Cs, I, K, Sr, Th, U and Zn by the adult population of Republic of Korea is given in Table 32. As may become evident from results, there are large intracountry variations for I as well as U, but the variations for other elements were rather small.

# 7.8.3. Organ content study

Human tissue samples of bone, liver, thyroid, muscle, lung and kidney were collected from 6 subjects who died of road accident and were certified by medical examiners to be in normal health prior to their death.

The results on contents of elements of importance in radiological protection for a number of organs are shown in Table 33. The variations in the contents of elements such as, Th and U in lungs, liver and even in the skeleton were almost an order of magnitude. The country laboratory generated data on the organ contents for some additional trace elements such as Na, K and Cl, which were not included in the scope of the CRP, and were therefore not covered by this report.

Element	No. of samples	Range	Mean	S.D.	Median
Ca (g)	5	0.38-0.75	0.51	0.15	0.43
Cs (µg)	5	2.45-6.82	4.86	1.90	5.74
I (μg)	4	53.6-527.3	188.7	227.5	86.98
K (g)	5	1.63-4.07	2.79	0.88	2.86
Sr (mg)	5	0.96–4.24	2.07	1.34	1.64
Th (µg)	5	0.78–1.47	1.10	0.28	1.00
U (µg)	5	0.36–10.37	5.18	4.86	3.80
Zn (mg)	5	6.87–12.89	9.82	2.19	9.52

TABLE 32. DAILY DIETARY INTAKES OF IMPORTANT ELEMENTS BY ADULTPOPULATION OF REPUBLIC OF KOREA

TABLE 33. ELEMENTAL CONTENTS OF SOME IMPORTANT ORGANS IN THE REPUBLIC OF KOREA

Element and Units	Organ	Content of various trace elements in human organs					
		No. of samples	Range	Mean (SD)	Median		
Cs (µg)	Muscle	6	468.0–919.5	633 (161.3)	623.6		
I (mg)	Thyroid	6	4.36-24.0	12.1 (7.0)	12.0		
Sr (mg)	Skeleton	6	224.6-733.5	436.7 (187.8)	412.0		
Th (µg)	Skeleton	5	4.2–38.0	23.8 (13.9)	22.1		
Th (µg)	Liver	6	0.00-0.33	0.20 (0.10)	0.23		
Th (µg)	Lungs	6	0.48-4.11	2.10 (1.50)	2.01		
U (µg)	Skeleton	6	2.84-29.03	9.45 (9.92)	5.23		
U (µg)	Liver	3	0.14-1.15	0.53 (0.54)	0.28		
U (µg)	Lungs	6	0.24–2.35	1.27 (0.81)	1.15		
U (µg)	Kidney	6	0.06-0.30	0.15 (0.08)	0.14		

# 7.9. Vietnam

#### 7.9.1. Ingestion study

In Vietnam, estimations of only dietary intakes of trace elements were carried out. Samples of duplicate diets were collected from four different regions of Vietnam namely, Hanoi, Ho Chi Minh City, Vinhphu and Dalat. The diet mainly consisted of ready to eat food items collected from public eating houses, food shops and canteens etc. Five samples of total diet were also prepared. The total diet samples (cooked), included 22–24 items as raw materials, which were washed and then cooked in typical Vietnamese style which meant boiling, frying, grilling and stewing of the food materials. In all,

18 diet samples of both the duplicate and total cooked diet were collected. The samples were homogenised and dried in oven at  $50-60^{\circ}$ C for 5–6 days and finally ground into powder. INAA and RNAA were employed for determining the concentrations of most of the trace elements in diet samples. For Th and U however, the analysis was carried out at NIRS, using ICP-AES and ICP-MS.

#### 7.9.2. Daily dietary intake

The dietary intakes of 22 elements were estimated for the adult population of Vietnam. The elements included in the study were Cs, I, Sr, Th, U, Ca, K, Mg, Na, P, Cu, Fe, Mn, Zn, As, Cd, Hg, Pb, Ba, Co, Cr and Ni. Although the results of the study of the toxic and some other elements studied only in Vietnam are included in the data table, they are not discussed further in this report. Results on daily intakes for the Vietnamese population are shown in Table 34 and include range, mean ( $\pm$ SD) and median intakes. The marked feature of the results is the extremely high intake of I by the Vietnamese population (discussed later in section 8.1.1). Surprisingly the difference between the median and the mean intake values for essential elements Fe, Cu and Zn is quite large indicating skewed distribution even for the essential trace elements.

Element	No. of samples	Range	Mean	S.D.	Median
Cs (µg)	18	5.85-20.3	9.61	3.83	8.96
I (µg)	18	0.6–2.4	1430	480	1449
Sr (mg)	18	0.13–7.15	1.26	0.79	1.29
Th (µg)	18	0.39–5.73	1.09	0.80	1.04
U (µg)	18	0.09–2.33	0.85	0.63	0.74
Ca (g)	18	0.06-1.44	0.66	0.42	0.64
K (g)	18	0.4–2.72	1.44	0.68	1.28
Mg (g)	18	0.07-0.238	0.123	0.052	0.13
Na (g)	18	0.53-4.55	2.91	1.5	2.76
P (g)	18	0.23-1.15	0.68	0.30	0.73
Cu (mg)	18	0.28–2.62	0.87	0.51	0.68
Fe (mg)	18	2.22-25.1	6.62	2.46	7.41
Mn (mg)	18	1.50-6.76	3.01	1.52	2.76
Zn (mg)	18	3.72-6.03	4.92	0.17	5.21
Cr (µg)	18	90–310	190	80	195
Co (µg)	18	4.32-18.95	9.56	3.79	10.13
As (µg)	18	33.8-362.3	91.67	34.6	94.5
Cd (µg)	18	7.8–51.0	16.64	11.28	11.8
Hg (µg)	18	3.655.82	4.98	0.71	5.15

TABLE 34. DAILY DIETARY INTAKES OF TRACE ELEMENTS FOR ADULT POPULATION OF VIETNAM

#### 7.10. Data presentation in box and whisker plots

The data on daily dietary intakes and organ contents of elements in populations of various Asian countries show very large inter-country and intra-country variations. Many of the data sets under consideration do not conform to a simple Gaussian (normal) statistics. It was therefore decided to use a method of data evaluation that is relatively insensitive to the exact form of the statistical distribution and to the presence of outliers. The method chosen was based on the use of the median and the range of values. These data are presented in the form of box and whisker plots. The box plots shown (Figs 1–24) illustrate the 75<sup>th</sup> percentile, the median, and the 25<sup>th</sup> percentile along with the minimum and maximum range of values obtained after the rejection of suspected outliers (beyond 1.5 R on both sides of the boxes 25<sup>th</sup> percentile and 75<sup>th</sup> percentile values), which are not shown in the range of values. The box encloses 50% of the data. The whiskers extend from the 75<sup>th</sup> percentile on the higher side and 25<sup>th</sup> percentile on the lower side until the end of the maximum and minimum range of acceptable values. As stated above, the outliers (unacceptable values), although included in the range of results in Tables, are not included in the box and whisker plots. Also shown in the box plots are the notches, which display the uncertainties of the medians. The size of the notch is 1.7 s on either side of the median, where

$$s = \frac{0.926 \text{ R}}{\text{N}^{1/2}}$$

R is the inter-quartile range (the difference between  $75^{th}$  and  $25^{th}$  percentiles) and N is the number of observations.

#### 8. DISCUSSION

Nine Asian countries, Bangladesh, China, India, Indonesia, Japan, Pakistan, Philippines, Republic of Korea and Vietnam participated in the CRP dealing with intakes and organ contents of Ca, Cs, I, K, Sr, Th and U, elements of importance in radiation protection. These elements have chemical and biological similarity with radionuclides <sup>137</sup>Cs, <sup>131</sup>I, <sup>90</sup>Sr, <sup>232</sup>Th, <sup>238</sup>U encountered during the production of nuclear electric power. Availability of accurate data on their biokinetic parameters could ensure realistic assessment of internal radiation dose to the humans. The data on these parameters such as the gastrointestinal absorption factor (f<sub>1</sub>), its uptake in human organs (f<sub>2</sub>) and its biological half-life or rate of clearance from the body (t<sub>b1/2</sub>), have been obtained from intakes and organ contents of the above mentioned elements by a number of workers [12, 15–17]. In addition, these data are useful to test the new ICRP models of these elements proposed by ICRP for global relevance in radiological protection.

Among the five radionuclides, <sup>137</sup>Cs, <sup>131</sup>I and <sup>90</sup>Sr are the most abundant fission products, which are released in nuclear fission at the time of nuclear reactor operation and also during a nuclear accident. <sup>232</sup>Th and <sup>238</sup>U are two long-lived naturally occurring radionuclides which are used as the nuclear fuel. The importance of Cs, I, Sr, Th and U is because they are stable (or near stable) isotopes of the above radionuclides. Two other elements included in the study: Ca and K are similar in chemical and biological behaviour to Sr and Cs, respectively, and therefore to their respective radionuclides <sup>90</sup>Sr and <sup>137</sup>Cs. The intakes of Ca and K in diet are higher than the chemically similar trace elements Sr and Cs by three orders of magnitude and could therefore influence the absorption of their radioactive isotopes <sup>90</sup>Sr and <sup>137</sup>Cs (most abundant fission products).

Data on Cu, Zn, Mn, Fe, Na, Mg, P, etc., the elements of importance in nutrition were also obtained. The intakes of these elements were studied to establish baseline intakes data for adult Asian population. The role of trace elements in health and disease has been well studied and documented [58–64] and these data could be useful in the study of elemental deficient diseases. Further, it was

recently discovered that essential trace elements such as Fe, Mn, Se, Zn, etc. could protect body organs against the harmful effects of radiation (radiation damage). It was shown that when Fe, Mn, Se and Zn were consumed in adequate quantities by experimental animals, the biochemical markers, which generally get elevated after radiation dose (due to radiation damage), were not significantly elevated [22, 23, 24]. The baseline data intakes of essential trace elements could be useful in further studies related to their role in protection against harmful effects of radiation.

In Indonesia data collected was based on a small number of diet samples which were collected and processed without strictly following the protocols developed by IAEA. The quality control in analysis was also lacking. In all, four elements of radiological importance and Zn were investigated. Although the total diet samples were prepared based on the market basket survey of food consumption, instead of taking total food materials consumed during one day, only one-tenth food materials were taken for the preparation of the diets. This approach slightly deviated from the standard approach. The use of a fraction of the food materials consumed often leads to an approximation and subsequently error in the estimation. Again, the quality control aspect which was the hallmark of the CRP (e.g. analysis of duplicate diet in the country laboratories and at the CRL followed by comparison of the results to work out z-scores) was not carried out. Organ contents data were reported for Ca, K and Sr on only three randomly collected bone samples which could not represent the entire adult population of that big country. Therefore, although the data from Indonesia are reported in this report for the purpose of information value to the readers, they were not considered for evaluation.

The present study in general used well-developed and harmonized protocols for sampling, sample preparation, strict quality control and uniformity in data reporting, a feature exercised by all the participating laboratories. Highly sensitive analytical techniques such as ICP-MS, ICP-AES, INAA, ENAA and RNAA were employed to carry out elemental analysis for diet and tissue samples. In addition to these methods, AAS was employed for estimating intake of a few of the essential elements.

The analysis of Cs, I, Sr, Th, and U in diet and tissue samples was a big challenge for the CRP participants. These elements were present in samples at mg/kg and sub-mg/kg levels. Mutual collaboration and cooperation between the participating laboratories and also with the CRL ensured reliable results. This was made possible because of the opportunity provided by the IAEA for the exchange programme at the level of experts as well as at the level of working scientists.

A highlight of the quality control in the present CRP was the role of the CRL (see Section 6.2.1). It was for the first time that such a concept was put into practice. The NIRS laboratory at Japan acted as the CRL for this project and ensured the external quality control. Simultaneous duplicate analysis was carried out on actual diet and tissue samples (10% of the total diet samples) at country laboratories as well as at the CRL and these results were used for calculating z-scores. The concept of z-score has already been discussed earlier in the quality control chapter (section 6.2.). The responsibility of internal analytical quality control was, however, left to the individual country laboratories following their participation in analyzing selected sets of RMs.

All Asian countries studied the daily dietary intake of elements of radiological importance. In the case of nutritional elements however, they used their discretion for the elements to be studied based on the availability of adequate analytical capabilities with them to analyse dietary samples. The number of elements studied in various countries ranged from 5 for Indonesia to 22 for Vietnam (see Table 16). China, Pakistan and Vietnam also studied intakes of toxic elements As, Hg, Pb, Cd and Sb. The data on the toxic elements were, however, not included in the present document for reasons stated earlier.

Diet samples were analysed by all participants while human tissue samples were analysed only in China, India, Republic of Korea and to some extent in the Philippines. In Indonesia, only three bone samples were analysed for Ca, K and Sr. A major problem associated in the collection of tissue

samples in the Muslim countries has been the social and cultural factor forbidding human autopsy. Therefore, no tissue samples were collected or analysed for the organ content in such countries. Even from Japan and Vietnam no data on organ content were reported.

The evaluation of both intakes and organ contents data on various trace elements was carried out to propose intake and organ content values representative of the Asian population. These values were compared with values of intake and organ content in radiation protection and nutrition bodies (see section 8.1).

# 8.1. Diet study

The diets were prepared by the market basket approach in most countries with the exception of Japan where all the samples were collected and prepared only by duplicate diet approach. A few additional duplicate diet samples were also collected even in all those countries where the market basket study approach was adopted. The aim was to capture the variability in the trace element intakes in these countries due to different dietary habits among various sections of its population. The extent of variability in the intake of trace elements has been very well depicted by data represented as box plots. They clearly show the median and range of intakes in individual Asian countries. It is also possible to get a clear idea of the inter-country variations in the intakes of elements from the box plots (see Figs 1-14).

About 2,800 determinations were performed on diet samples in nine Asian countries for up to 22 elements. The details of analytical methods used, the elements analysed and the diets studied in each individual country have already been described earlier (Table 2).

In this document, the discussion is limited to only two groups: elements of importance in radiological protection Ca, Cs, I, K, Sr, Th, U, and elements of importance in nutrition, namely, Cu, Fe, Mn, Mg, Na, P and Zn. Details are given in Table 16. Since large intra-country variations were observed in the intakes of most elements, it was decided to use mainly the median values instead of the means for realistic representation of data. However, the ranges, means and standard deviations are also reported in the data tables to give an idea of the variability of the data.

Additional essential and toxic elements, namely Cr, Co, Se, Ba, Ni, Pb, Cd, Hg, As and Sb, were studied in some of the participating countries but have not been included in this document.

The data from the Asian region were compared (Table 35) with intake values found in ICRP reference man [3], the RDA values proposed by the US National Academy of Sciences [79], and also with the global intake values compiled by the IAEA [86]. While making a comparison of essential elements, it is important to remember the significantly lower body weight for the adult Asian population [10] in comparison with ICRP-23, representing European and North American population. This is important since intake or requirement of an element or a nutrient may depend, among other factors, on body weight of the population, as dealt with elsewhere [87].

# 8.1.1. Daily dietary intakes of trace elements of importance in radiation protection

# Caesium

The daily dietary intakes of Cs were studied in all participating Asian countries. The intakes showed variation by a factor of five (Table 36). The lowest intake of 4.35  $\mu$ g was reported from India and the highest value of 23.6  $\mu$ g was from Bangladesh. The median intake of Cs for Asian populations was

6.5  $\mu$ g which is somewhat lower than the ICRP-23 value of 10  $\mu$ g. The lowest intake of Cs in India and its overall lower intake in the Asian region could be attributed to the lower intake of milk, its products and flesh foods. A comparison of the intakes of milk and other food materials consumed by populations of Europe (and North American countries) and Asian countries is shown in Table 37. As can be seen from the table, the intakes of milk, milk products and also flesh foods consumed in the Asian region are very low in comparison to the intake by Caucasian population. A clear idea on the distribution of daily dietary intakes of Cs in Asian countries could also be obtained from the box plots (Fig. 1). The data on dietary intake and median organ content of Caesium could be applied for population in the Asian region to theoretically derive the expected biological-half life of <sup>137</sup>Cs (fission product).

Element	Asian Values		ICRP-23	Global Average Value	US RDA
	Range	Median	[3]	[86]	[79]
Ca (g)	0.22-0.72	0.45	1.10	0.76	0.8
Cs (µg)	4.35-23.58	6.5	10.0	8.80	
I (μg)	40.0–1449.0	90.0	200	190	150
K (g)	1.0-2.86	1.9	3.30	2.67	1.9–5.6
Sr (mg)	1.36–2.88	1.6	1.9	1.50	
Th (µg)	0.15-3.53	1.0	3.00	1.40 (*)	
U (µg)	0.54–5.04	1.0	1.90	1.20(*)	
Cu (mg)	0.68–1.95	1.1	3.50	1.50	2.0-3.0
Fe (mg)	4.22–30.1	11.0	16.0	13.0	12.0
Mn (mg)	2.34-11.75	3.5	3.70	3.10	2.5-5.0
Mg (mg)	0.13-0.52	0.3	0.34	0.30	-
P (g)	0.56-0.73	0.75	1.40	1.30	0.7
Na (g)	1.88-4.13	3.5	4.40	3.51	1.1–3.3
Zn (mg)	5.21-14.25	8.7	13.0	10.0	15.0

# TABLE 35. A COMPARISON OF PROPOSED DAILY INTAKES OF ELEMENTS FOR ASIAN POPULATION WITH ICRP AND GLOBAL VALUES

(\*) Global intake of Th and U intakes reported by UNSCEAR [68].

Country	Ca (g)	Cs (µg)	Ι (μg)	K (g)	Sr (mg)	Th (µg)	U (μg)
Bangladesh	0.31	23.58	-	-	1.83	3.53	1.31
	0.25-0.35	11.01-40.75			1.02-2.56	1.33-16.99	0.51-8.18
China	0.72	12.98	327	2.09	2.88	3.13	5.04
	0.62-0.91	10.33-15.51	175.2-631.9	1.69-2.56	1.99-3.67	1.14-14.07	1.56-15.36
India	0.36	4.35	85.0	1.57	1.50	0.64	0.54
	0.17-0.67	2.64-11.8	56.9-260	1.24-3.84	0.76-2.96	0.44-1.75	0.35-1.16
Japan	0.44	7.01	246	1.91	1.60	0.32	0.64
	0.12-1.29	3.51-19.51	38.0-8710	1.15-2.62	0.52-3.86	0.12-0.97	0.13-6.52
Republic of Korea	0.43	5.74	87.0	2.86	1.64	1.00	3.80
	0.38-0.75	2.45-6.82	53.60-527.3	1.63-4.07	0.96-4.24	0.78-1.47	0.36-10.37
Pakistan	0.48	5.97	43.5	2.62	2.43	2.66	2.16
	0.22-0.71	2.60-12.90	15.4-139.2	1.70-4.30	0.91-5.70	0.92-7.15	1.35-6.70
Philippines	0.22	5.25	40.0	1.01	1.36	0.15	0.56
	0.11-0.69	2.56-28.13	21.5-519.4	0.55-1.81	0.69-5.15	0.02-1.69	0.11-4.82
Vietnam	0.64	8.96	1449	1.28	1.29	1.04	0.74
	0.06-1.44	5.85-28.3	600-2400	0.4-2.72	0.13-7.15	0.39-5.73	0.09-2.33
Asian Range	0.22-0.72	4.35-23.58	40-327*	1.1-2.86	1.36-2. 88	0.15-3.53	0.54-5.04
Asian Median (rounded)	0.45	6.5	90	1.90	1.6	1.0	1.0

# TABLE 36: DAILY INTAKES OF THE ELEMENTS OF IMPORTANCE IN RADIATION PROTECTION IN A FEW ASIAN COUNTRIES (MEDIAN VALUES AND RANGES)

\* Excluding Vietnam (see discussion).

TABLE 37: COMPARISON OF THE DAILY INTAKES (g) OF PRINCIPAL FOOD COMPONENTS IN REGIONS REPRESENTED BY CAUCASIAN POPULATIONS AND IN THE ASIAN REGION (adapted from references [3] and [10])

Food groups	UK	CEE	USA	Asian countries range
Milk	382	287	508	9–147
Cheese	12	21	19	-
Total flesh foods	158	140	228	12-172
Meat products	137	118	206	
Fish & other sea-foods	21	22	22	12-172
Eggs	34	21	47	2–46
Fats	44	63	49	6–33
Sugar	77	57	69	1–64
Total vegetables	320	376	305	65–324
Potatoes	202	196	103	-
Other vegetables	118	180	202	-
Fruits	108	114	184	3–165
Cereals	246	346	207	171–520
Pulses	-	-	-	35–114



FIG. 1. Daily dietary intakes of Cs by adult population in some Asian countries.

#### Iodine

The intakes of I were studied in 7 countries. The two exceptions were Bangladesh and Indonesia. Diet samples in Bangladesh were oven dried instead of freeze drying. The samples were not analysed for I because of the inadequate QC steps observed during sample preparation. For most of the countries, I analysis was carried out at a contract laboratory in Prague, Czech Republic. Indian and Vietnamese participants carried out I analysis in their own laboratories. Indian participants however used the analytical facility at Czech laboratory to confirm their results. A low daily intake 40 µg of I was observed in the Philippines, while a maximum of 1449 µg was reported in Vietnam, followed by Japan and China. Again, in this case, a large inter-country variation in the intakes by a factor of more than an order in magnitude was observed. The high intake of I in Japan was reportedly due to the consumption of marine algae, seaweeds and other marine food materials which are rich in I. The reason for extremely high intake of I in Vietnam could not be exactly identified but most likely it is the use of iodised salt with excess of iodine in some diet samples. Another possible reason could be the consumption of seaweeds and other seafoods similar to those consumed by the Japanese population. Despite the very high intake of I reported in Vietnam, the median intake value of 90  $\mu$ g of I for the Asian region was low (Fig. 2) as the intakes in most of the other Asian countries were consistently low and the extreme value from Vietnam did not affect the median value.

The median intake of I in the Asian region was lower than both the ICRP-23 value of 200  $\mu$ g [3] and the global average intake of 150  $\mu$ g [86]. The reason for the lower intake in Asian region could again be traced to low consumption of milk and milk products which have higher concentrations of iodine in it. This aspect has been well brought out in Table 37. Whereas the daily intake of milk and milk products was about 500 mL for Caucasian population, Asian countries ranged between 9 and 147 mL. The box plots provide a clear picture of daily intakes for I in the Asian region (Fig. 2). The highest intake among five countries shown in the box plot is for Japan. The intake in Vietnam was mainly from the iodised salt and did not represent the natural dietary consumption of I by the Vietnamese population and was therefore not included in box plots.



FIG. 2. Daily dietary intakes of I by adult population in some Asian countries.

#### Strontium

Sr was considered an important element for this study since it is the stable counterpart of the betaemitting radionuclide and fission product <sup>90</sup>Sr. Daily intakes of Sr were studied in all nine countries. Unlike other elements discussed above, intakes of Sr in different Asian countries varied by a factor of <3. The minimum daily intake of 1.29 mg Sr was obtained for Vietnam and the maximum of 2.88 mg Sr was obtained for the population of China. The median intake of Sr for the Asian region was 1.6 mg, which was similar to the global average intake (Table 35) and was only marginally lower than the intake of 1.9 mg Sr, reported in ICRP-23. Interestingly enough, if one takes into account the 20% lower body weight of the Asian population [10] when compared to the Caucasian population, the Sr intake per kilogram of the body weight in Asian region would compare well with the global average and also with the ICRP-23. The box plots for the intake of Sr (Fig.3) clearly show the highest intake of Sr by the population of China and the lowest intake by the population of Vietnam.



FIG. 3. Daily dietary intakes of Sr by adult population in some Asian countries.

#### Thorium

Th is the source material for nuclear fuel (<sup>233</sup>U), having potential for its use in the next generation of nuclear power reactors (fast breeder reactors). Th itself is a fertile material which is converted into fissile <sup>233</sup>U, through the nuclear reaction <sup>232</sup>Th (n,  $\gamma$ ) <sup>233</sup>Th----B---<sup>233</sup>Pa---- B ---<sup>233</sup>U. The handling of Th and its compounds in its prospecting, mining, milling, separation, purification, fuel fabrication could lead to radiation exposure of workers. Th is also used in gas mantle industry. Intakes of Th were studied in 8 Asian countries except Indonesia. They varied by more than an order of magnitude. The highest daily intake was 3.5 µg in Bangladesh followed by 3.13 µg in China, while the lowest intake 0.15 µg was reported from the Philippines. The intakes are shown in Table 36. One possible reason for the extremely low intake of Th and also of a few other trace elements by the Filipino population could be the low food intake. The box plots clearly show the highest intake of Th in Bangladesh with equally large intra-country variations and the associated uncertainty with the median as indicated by the notch extending below the 25<sup>th</sup> percentile level (Fig. 4).



FIG. 4. Daily dietary intakes of Th by adult population in some Asian countries.

The median intake of Th for the Asian population was obtained as 1  $\mu$ g which is 3 times lower than the ICRP-23 value (Table 36). Its intake reported for the New York residents by Fisenne et al. [65] was also 1  $\mu$ g, and similar to the Asian intake. Shiraishi et al. [66] reported somewhat lower daily intake of 0.42  $\mu$ g in the Japanese population, estimated on the basis of the analysis of duplicate diet. Another set of workers also from Japan [67] estimated the daily intake of 0.68  $\mu$ g Th for the Japanese population based on market basket study of 18 food categories. Both Japanese values were however in the range of intake reported for the Asian countries.

The proposed Asian value is also similar to the global intake value of 1.2 µg Th as proposed by United Nations Scientific Committee for Effects on Ionizing Radiation (UNSCEAR) [68].

#### Uranium

U is an important fuel element which, in its natural form (as natural uranium) as well as enriched U, is used in different types of reactors. The workers involved in the prospecting, processing of the ore, in various stages of its purification and fuel fabrication could get exposed to this radioactive element. Both, <sup>238</sup>U, the main component of natural uranium, and the enriched uranium <sup>235</sup>U (fissile material) have very long half-lives (more than a billion years). Once a subject is exposed to them, the radioactive uranium isotopes could remain in the human body (in skeleton) for a fairly long time. U is also known for its chemical toxicity which becomes the deciding factor for limit on daily intake of its soluble compounds.

In regions of some Asian countries and also in other parts of the world (India, Canada, China, Republic of Korea), concentration of U in soil as well as in the ground water is reported to be significantly high. The drinking water if used from these regions for cooking and drinking purposes (as part of the fluids), could significantly increase its daily intake. Such regions cover only a small area of total Asia and it was therefore decided at the beginning of the project itself to exclude diet samples from these regions.

Except Indonesia, the intakes of U were estimated in all other Asian countries. The inter-country variations in the daily intakes were observed to be quite large and could become clear from the box plots (Fig. 5). The minimum intake of 0.5  $\mu$ g was reported from India while the highest of 5.0  $\mu$ g was reported for the population of China. The Asian median of 1  $\mu$ g U is half of 1.9  $\mu$ g cited in ICRP-23 (Table 36) but it is similar to the daily intake of 1.2  $\mu$ g U reported by Fisenne et al. [65] for residents of New York. Two Japanese groups [66, 67] reported U intakes of 0.73  $\mu$ g and 1.2  $\mu$ g U by the Japanese population. Although the Asian median intake is lower, it is similar to the US subjects from New York City and also comparable to the global average of 1.3  $\mu$ g recommended by UNSCEAR. These data are useful for future revision of the ICRP intake value of U.



FIG. 5. Daily dietary intakes of U by adult population in some Asian countries.

#### Calcium

Ca has a dual role of being important in radiological protection and also as an essential element. Ca is similar to Sr in chemical behaviour and therefore bears metabolic relationship to the important fission product <sup>90</sup>Sr. This is also an essential and structurally important element for the human system. As calcium phosphate, it forms an integral part of the human skeleton. Because of similarity in behaviour with Sr, the extreme changes in the intake of Ca had been reported to effectively influence the intake of <sup>90</sup>Sr [19].

The two main sources of Ca in diet are milk and vegetables. In the present study it was observed that consumption of these food materials is much less by the Asians than by the Europeans and North Americans. Table 37 compares consumption of some major food components in Europe, America and the Asian region. It becomes sufficiently clear from the Table that milk and green vegetables are consumed in relatively smaller quantities in Asia.

In Asia, the intake of Ca was highest in China, followed by Vietnam. The intake varied between 0.22 g for the Philippines to 0.72 g for the Chinese population. Similar to the trend observed for Sr, the daily intake of Ca in all of the Asian countries did not vary appreciably. A special feature of its intake in Vietnam was large intra-country intake variation. This feature becomes clear from the box plots (Fig. 6). The Asian median intake is 0.45 g Ca, which is significantly lower than 1.1 g Ca mentioned [3], 0.76 g for the global average [86] reported by IAEA (Table 35) and 0.8 g proposed for the US RDA [79]. From a radiological safety point view point, the possible effects of lower intake of Ca in certain Asian countries needs to be studied in the context of the absorption of <sup>90</sup>Sr.



FIG. 6. Daily dietary intakes of Ca by adult population in some Asian countries.

#### Potassium

Similar to Ca, the K also has a dual role. It is important from a radiological viewpoint, because of its chemical similarity to Cs and therefore to <sup>137</sup>Cs (the most abundant fission product). It is also an essential minor element for the human system. The element K in association with Na is responsible for maintaining the integrity of the human cell by maintaining the Na-K pump that controls and balances the intra and extra-cellular fluid in human body.

Except Bangladesh, all other countries studied dietary intakes of K. Unlike Th, U and Cs, the variations in intakes of K in different countries were not very large and ranged from 1.04 to 2.69 g per
day with the Asian median as 1.9 g. The lowest intake was reported from the Philippines and the highest from Pakistan. The possible reason for lower intake of K by the Philippine population was perhaps the intake of small quantity of food materials as explained earlier in section on intake of Th.

The intake pattern of K in the Asian region becomes clear when one observes the box plots for potassium (Fig.7). Median intake 1.9 g in Asian countries is lower than the 3.3 g reported in ICRP-23 and also lower than the global average value of 2.87 g. From the viewpoint of nutrition and its availability through diet, its intake is comparable to its requirement as indicated by RDA value of 1.9 g for the 73 kg male (average weight for Caucasian male) subjects.



FIG. 7. Daily dietary intakes of K by adult population in some Asian countries.

# 8.1.2. Daily dietary intakes of trace elements of importance in nutrition

A comparison of daily dietary intakes of essential trace elements from nine Asian countries is shown in Table 38. Results for intakes of various essential elements are discussed below.

# Iron

Iron is an important trace element whose deficiency can lead to iron deficiency anaemia. Its daily dietary intakes were studied in 8 countries and they ranged from 5.4 mg for Japan to 30.3 mg for Pakistan. The results are shown in Table 38. The higher intake in Pakistan is perhaps due to large consumption of wheat (staple food of the population) which is rich in Fe. The intakes by the populations in various countries are well represented in box plots (Fig. 8). The intakes vary by a factor of five within the Asian region. Daily intakes of Fe in some Asian countries, for example Japan, are quite low in comparison with the ICRP-23 value of 16.0 mg. On the other hand, when comparing the average body weight of 73 kg of Caucasian population with the average body weight of about 60 kg for the Asian population, the Asian median intake of 11 mg Fe may not appear significantly low in comparison with the US RDA value of 12 mg.

Country	Cu (mg)	Fe (mg)	Mn (mg)	Mg (g)	P (g)	Na (g)	Zn (mg)
Bangladesh	1.95 0.68-2.23	10.80 8.30-18.02	3.38 2.01-4.08	0.32 0.21-0.48	-	-	9.07 3.92-12.37
China	1.39 1.29-2.31	30.10 16.90-44.90	6.67 5.36-8.58	0.36 0.26-0.45	-	3.75 1.96-5.11	12.01 11.66-14.22
India	-	14.85 10.20-34.40	-	-	-	-	7.86 5.3-15.60
Japan	1.06 0.44-1.73	4.91 2.80-15.87	3.51 1.34-5.45	0.21 0.12-0.27	0.79 0.52-1.26	4.13 2.07-5.27	8.27 4.66-12.75
Rep. of Korea	-	-	-	-	-	-	9.52 6.87-12.89
Pakistan	-	30.03 12.46-51.61	11.75 3.70-24.36	0.52 0.22-0.92	-	3.43 3.32-3.65	14.25 7.62-23.10
Philippines	0.95 0.66-3.71	4.22 2.44-43.84	2.34 0.95-6.76	0.13 0.06-0.21	0.56 0.30-0.84	1.88 1.22-2.78	5.23 2.35-8.17
Vietnam	0.68 0.28-2.62	7.41 2.22-25.1	2.76 1.50-6.76	0.13 0.07-0.24	0.73 0.23-1.15	2.76 0.53-4.55	5.21 3.72-6.03
Asian Range	0.68-1.95	4.22-30.10	2.34-11.75	0.13-0.52	0.56-0.79	1.88-4.13	5.21-14.25
Asian Median (rounded)	1.1	11.0	3.5	0.30	0.75	3.5	8.5

# TABLE 38: DAILY INTAKES OF THE ELEMENTS OF IMPORTANCE IN NUTRITION IN A FEW ASIAN COUNTRIES (MEDIAN VALUES AND RANGES)

Source of information in the above table: ICRP reference Man Data, ICRP-23 [3]

(\*) Data extracted from the final report on phase-1 of the reference Asian Man Project [10]

(1) Assumes 10% waste from those purchased

(2) Actual consumption

(3) Assumes 15% waste from those purchased.



FIG. 8. Daily dietary intakes of Fe by adult population in some Asian countries.

Zinc

Zn is an essential trace element for the growth as well as maintenance of the human system. It is a cofactor in a number of metallo-enzymes. Carbonic anhydrase, alcohol dehydrogenase, and alkaline phosphatase are a few examples of Zn enzymes. Its deficiency is reported to be associated with hypogonadism, dwarfism, and the reduction of body's auto-immune response. The daily intakes were estimated in all participating Asian countries. The comparative intakes in different Asian countries are shown in Table 38. The daily intakes varied from 5.21 mg for Vietnam to the highest intake of 13.6 mg for the population of Pakistan. Although the intake appears high in Pakistan among the participating countries, even this intake is just comparable to the nutritionally defined requirement value of 15 mg per day. The possible reason for the adequate intake of Zn in Pakistan is the consumption of a large quantity of wheat (the staple food). Wheat has high concentration of Zn, Fe and Mn in comparison with their relatively low concentration in rice. Only in Pakistan and China, the intakes were adequate. In the Philippines, Vietnam, India and even in Japan, the intakes were significantly lower than the US RDA for Zn and also much lower than 15 mg cited in ICRP-23 [3]. It is interesting to note (see Table 38) that the Asian median intake of 8.5 mg Zn (Fig. 9) is not much lower than the global average intake value of 10.0 mg (see Table 35).



FIG. 9. Daily dietary intakes of Zn by adult population in some Asian countries.

#### Manganese

Intake of Mn was studied in six Asian countries. The intakes varied from a minimum of 2.34 mg for the Philippines to maximum intake of 11.75 mg for the population of Pakistan. The highest intake of Mn by the population of Pakistan is also due to the consumption of a large quantity of wheat which is rich in Mn. The Asian median intake was found to be 3.5 mg. Mn is one of the few essential elements whose intake in Asian population was found to be similar to the value as seen from ICRP-23 [3] and US RDA (Table 35). Except for the population of Pakistan, the inter-country variations for the intake of Mn in various Asian countries is not very high as may become clear from the box-plots (Fig.10).



FIG. 10. Daily dietary intakes of Mn by adult population in some Asian countries.

#### Copper

Cu is an essential trace element and is associated with the metallo-protein, ceruloplasmin present in blood. Its deficiency is known to cause secondary anaemia. It has also been recognized as a co-factor in a number of other metallo-enzymes such as cytochrome-c-oxidase, superoxide dismutase etc. Its intakes were studied in only five Asian countries. The range of daily intakes varied from 0.87 mg for Vietnam to 1.8 mg for Bangladesh. The median intake of 1.1 mg for Cu in the Asian region is much lower than the ICRP-23 value. It is also low when compared to US RDA value of 2.0–3.0 mg (Table 35). The variation in the intake of Cu in the Asian region was observed to be small as may be seen from box-plots for the intake of copper (Fig. 11).



FIG. 11. Daily dietary intakes of Cu by adult population in some Asian countries.

#### Magnesium

Magnesium is an essential minor element. Its deficiency is known to be associated with heart disease. The intake of Mg was studied in six Asian countries. Intakes varied by a factor of 5. As may be seen from Table 38 and Fig. 12, the minimum intake of 0.13 g was for the Philippine population and the maximum of 0.52 g was for Pakistan. The median intake of 0.30 g was comparable to ICRP-23 value of 0.34 g. No RDA value is available for comparing intake of Mg in the Asian region.



FIG. 12. Daily dietary intakes of Mg by adult population in some Asian countries.

#### Phosphorus

Phosphorus along with Calcium, belongs to the category of structural elements and forms an integral part of the human bone (skeleton). As may be seen from Table 38, only three countries studied the intake of P, which varied by a very small factor (<1.5). The Asian median intake of 0.75 g phosphorus (Fig. 13) was much lower than 1.4 g mentioned in ICRP-23, but it was comparable to the US RDA value of 0.7 g. It was, however, lower than the global average value of 1.3 g.



FIG. 13. Daily dietary intakes of P by adult population in some Asian countries.

### Sodium

The main source of intake of Na is common salt used for cooking purpose. Only four countries studied its intake. It ranged between 1.88 g for Philippines and 4.13 g for Japan. The intake of Na in Pakistan was found to be 3.43 g. The Asian median intake was 3.5 which, although less than ICRP-23 value of 4.4 g, is adequate according to US RDA. The intakes pattern of Na in various Asian countries is shown in the Fig. 14.



FIG. 14. Daily dietary intakes of Na by adult population in some Asian countries.

# 8.1.3. Daily dietary intake of toxic and other elements

The daily dietary intake of toxic trace elements As, Sb, Pb, Cd and Hg, and a few other essential elements such as Se, Cr, Co Ni, Ba, etc. were also studied in some of the Asian countries. As they belonged to the category of optional elements (of interest to the individual country), the number of studies reported for most of these elements ranged from 1 to 3 and therefore proper inter-country comparison of the results was not possible. The data on these elements were therefore not included in the present report.

# 8.2. Organ content of elements of importance in radiological protection

The contents of Cs, I, Sr, Th, U were determined in selected human organs where these elements are known to deposit in higher concentrations. The data on organ contents along with data on daily intakes of trace elements can be employed: 1) to estimate their biological half-lives, 2) to study their uptakes by various human organs, and 3) to appraise (test) some aspects of currently proposed ICRP models. The organ contents study was carried out only in India, China, Philippines and Republic of Korea. Very limited data obtained from Indonesia were not considered for evaluation (see section 8). The organ contents study could not be carried out in Bangladesh, Pakistan and Indonesia for reasons stated earlier. It is believed that data produced in neighbouring countries with similar physical traits and lifestyle, could still be effectively used in these three countries for generating the required radiological protection standards. An overview of the elemental contents of various organs reported from different countries is given in Tables 39 and 40. Individual organs are discussed in more detail below.

# Iodine in thyroid

The contents of iodine in thyroid were estimated for the population of China, India and Republic of Korea. Median values ranged from 6.27 mg for the Indian population to 19.34 mg for the Chinese population (Fig.15). Intra-country variations in the I contents of up to more than 5 times were observed. It is interesting to note that the dietary intake of I for Indian population was lower than that for Chinese population and correspondingly, the content of I in the thyroid of Indian population was also observed to be lower in almost the same ratio. The Asian median of 12 mg (Table 39) was the same as in ICRP-23.

Organ	Element	China		India		Republic	Asian value (ICRP-23 value)	
		Range (n)	Median content	Range	Median	Range	Median	
Thyroid	I (mg)	6.61-36.88 (24)	19.34	4.36-12.6 (14)	6.27	4.36-24.0 (6)	12.0	12.0 (12.0)
Kidney	U (ug)	0.01-1.10 (33)	0.26	0.08-0.48	0.19	0.06-0.30	0.14	0.19 (7.0)
Liver	U (ug)	0.25-7.27 (21)	0.77	0.03-0.15 (21)	0.08	0.14-1.15 (3)	0.28	0.20 (0.45)
Liver	Th (µg)	0.22-6.53 (19)	0.53	0.09-0.23 (9)	0.12	0.00-0.33 (6)	0.23	0.23 (-)
Lungs	Th (µg)	0.34-50.18 (33)	7.79	1.2-9.68 (23)	4.35	0.48-4.11 (6)	2.01	3.2 (-)
Lungs	U (µg)	0.29-10.18 (33)	1.92	0.5-2.64 (15)	1.02	0.24 <b>-</b> 2.35 (6)	1.15	1.1 (1.0)
Skeletal Muscle	Cs (µg)	134.0 <b>-</b> 1870.0 (33)	469.0	153.2-669.8 (10)	327.0	458.0-666.2 (6)	623.6	469 (570)
Skeleton	Th (µg)	2.12-137.64 (32)	14.45	1.29-12.9 (19)	3.96	4.2 <b>-</b> 38.0 (6)	22.1	14.4 (-)
Skeleton	U (µg)	4.67-70.7 (33)	12.58	0.65-12.1 (15)	2.85	2.84-29.03 (6)	5.23	5.2 (59.0)
Skeleton	Sr (mg)	6.43-1068.8	353.15	42.5-580.0 (11)	172.5	224.6-733.5 (6)	412.0	353 (320)

TABLE 39: ORGAN CONTENTS OF SOME TRACE ELEMENTS OF IMPORTANCE IN RADIOLOGICAL PROTECTION IN ASIAN POPULATIONS

# TABLE 40. ORGAN CONTENTS OF SOME TRACE ELEMENTS OF IMPORTANCE IN RADIOLOGICAL PROTECTION IN ASIAN POPULATIONS (cont.)

Organ	Element	Unit	Philip	pines	Proposed Asian Values (ICRP-23 value)
			range	median	
Thyroid	Ι	mg	-	-	12.0 (12.0)
Kidney	U	μg	-	-	0.19 (7.0)
Liver	U	μg	0.04–0.21 (10)	0.12	0.20 (0.45)
Liver	Th	μg	0.00-0.33	0.23	0.23 (-)
Lungs	Th	μg	0.43-2.10	0.89	3.2 (-)
Lungs	U	μg	0.21-0.61	0.36	1.1 (1.0)
Skeletal Muscle	Cs	μg	-	-	469 (570)
Skeleton	Th	μg	-	-	14.4 (-)
Skeleton	U	μg	-	-	5.2 (59.0)
Skeleton	Sr	mg	-	-	353 (320)



FIG. 15. Contents of I in thyroid in adult population of some Asian countries.

Shortly after completion of the project described in this report, ICRP issued some new recommendations in 2003 on basic anatomical and physiological data for use in radiological protection. This new document, Publication 89, is discussed in more detail in Section 11 (Addendum). It addresses briefly the issue of iodine in the thyroid, quoting a value of 0.1% by mass.

As discussed in more detail in the Addendum, this value is compatible with the results of the present IAEA study in view of the rather wide confidence intervals that apply to both sets of estimates.

### Uranium in different human organs

The uranium contents were estimated in kidneys, liver, lungs and skeleton in China, India and Republic of Korea. A limited amount of data on U in lungs and liver were also collected in the Philippines.

# Uranium in kidney

The uranium entering the human body is deposited in highest concentration in the kidneys. The safe limit of its daily intake (soluble form) is also governed by the fraction of ingested uranium that deposits in the kidneys. If consumed in higher quantity, U is known to damage kidneys. The data from Asian countries on the U contents of human kidneys were therefore important from the viewpoint of its chemical as well as radiotoxicity. Due to non-availability of reliable analytical methods used in earlier years, very high content of U in the kidneys (7  $\mu$ g) was reported for subjects living even in normal background area and having no specific history of exposure to this element [13]. Relatively recent studies from the USA, India and Japan have reported about 30 times lower content of U in the kidneys [69–71].



FIG. 16. Contents of U in kidneys in adult population of some Asian countries.

The contents of U in kidneys were estimated in China, India and Republic Korea. They ranged from 0.14  $\mu$ g for Republic of Korea to 0.26  $\mu$ g for the Chinese population with an Asian median of 0.19  $\mu$ g (Table 38). In a study reported from Japan [71] around the same time as the duration of this CRP, kidneys content of 0.13  $\mu$ g was obtained for the Japanese population. This value is close to the range of values obtained from other countries for the Asian population. It is interesting to note that there is very little variation in data reported from three Asian countries (Fig. 16). Another interesting observation was of the highest content of U in kidney in the Chinese population. The U intake for this population was also the highest among all Asian countries (Table 35).

The Asian median value of U in kidney was about 40 times lower than the 7.0  $\mu$ g cited in Ref [3]. This observation underscores the need to review U in kidney values. The implication of the lower organ content value on the biokinetic model may also need to be re-examined.

# Uranium in liver

Liver is not an important organ for the deposition of U, but it is regarded as a general storage organ for most trace elements, especially heavy toxic elements. Hepatic cells tend to filter out toxic elements from body fluids and in that process tend to store them. The contents of U in liver were studied in China, India, Republic of Korea and the Philippines. The content ranged from 0.08  $\mu$ g for the Indian population to 0.77  $\mu$ g for the Chinese population, with Asian median of 0.20  $\mu$ g (Table 39). The highest content of U in liver of the Chinese population is also indicated from box plots (Fig. 17). It is consistent with the elevated daily intake by the Chinese population. The 0.38  $\mu$ g U content of liver obtained for the Japanese population [71] is within the range of values obtained in the CRP in other Asian countries.



FIG. 17. Contents of U in liver in adult population of some Asian countries.

# Uranium in lungs

The organ content of U in lungs is not directly related to the ingestion. In fact only a small fraction of what is ingested may appear in lungs. The main source of U deposited in lungs is inhalation (dust). The soil particles that get airborne as dust contain as high as  $2-3 \ \mu g/g$  of U in them. They get deposited in the pulmonary region of lungs following inhalation. Since U in the air/dust is insoluble, it remains in the pulmonary region of lungs for tens of years [72].

The lung content of uranium was obtained in China, India, Republic of Korea and the Philippines. The highest value was reported for the Chinese population and is very well reflected in the box plots (Fig.18). Its contents in lungs ranged from 0.36  $\mu$ g for the Filipinos to 1.92  $\mu$ g for the Chinese (Tables 39 and 40). The Asian median value is 1.1  $\mu$ g. The Asian median is higher than 0.5 $\pm$  0.39  $\mu$ g reported by Fisenne and Welford [69] for the US population. The higher U content in the Asian region could be attributed to higher level of dust in the tropical Asian regions like India, etc.



FIG. 18. Contents of U in lungs in adult populations of some Asian countries.

#### Uranium in skeleton

Although the highest concentration of U is found in the kidney, because of the large mass, the largest quantity of U is present in the skeleton.

U in skeleton was estimated in India, China and Republic of Korea. Minimum value of 2.85  $\mu$ g was reported for the Indian population and the maximum of 12.58  $\mu$ g for the Chinese population. The higher content of U in skeleton of the Chinese population is again consistent with its highest intake. Igarashi *et al.* [71] reported the skeleton content of 7.5  $\mu$ g U for the Japanese population. This value lies within range of U content reported from other countries participating in the organ content study programme of the CRP. The contents of U between the three countries vary by a factor of about 5. The Asian median of 5.2  $\mu$ g was an order of magnitude lower than the ICRP-23 value of 59  $\mu$ g reported on the basis of data generated three decades ago by Hamilton [73]. This fact underscores the need for revision of reference on uranium content. The organ contents of the population of different Asian countries are shown in box plot (Fig. 19).

#### Thorium in human organs

The contents of Th were studied in liver, lungs and skeleton. The organs were selected on the basis of the ICRP-30 [13], indicating the organs with the maximum Th content. Whereas, skeleton and liver form parts of systemic compartment in the body, lungs, which contain the highest concentration of Th, are considered non-systemic. Th content of lungs represents the fraction deposited from inhalation and those of skeleton and liver represent contributions mainly from ingestion.



FIG. 19. Contents of U in skeleton in adult population of some Asian countries.



FIG. 20. Contents of Th in liver in adult population of some Asian countries.

#### Thorium in liver

The contents of Th in the liver were studied in China, India, Republic of Korea and the Philippines. The lowest concentration of Th was reported from India and the highest from China as is clear from the box plots (Fig. 20). The highest intake was also reported from China. The Asian median was found to be 0.23  $\mu$ g. For the Japanese population, 0.2  $\mu$ g Th in the liver reported using ICP-MS [74] was close to the Asian median. ICRP-23 has no data on Th in liver.

### Thorium in lungs

Th in lungs was studied in China, India, Philippines and Republic of Korea. The highest content of 7.79  $\mu$ g in lungs was reported for the Chinese population while it was only 0.89  $\mu$ g for the Philippines. The median content of lung for the population in the Asian region was 3.2  $\mu$ g, which compared well with the Th concentration of 0.32 pCi/kg (2.9  $\mu$ g/kg) reported by Ibrahim et al. [75] for subjects of Washington D.C. area of USA. Assuming weight of the lungs as one kilogram, this concentration will correspond to about 2.9  $\mu$ g of Th. The results are reported in Tables 39 and 40. The pattern of Th in lungs in various Asian countries also becomes clear from box plots (Fig. 21). Some very high Th content values were reported from China. Sampling lung tissue can be critical especially for Th. If while sampling the high thorium containing lymph node area of the lung is collected along with the normal lung tissue, then there could be false elevation of Th content in lung. It is therefore important that the lymph nodes are avoided during collection of the lung samples. Jaiswal et al. have reported more than ten times higher concentration of Th in pulmonary lymph nodes in comparison with the rest of the lung tissue [76].



FIG. 21. Contents of Th in lungs in adult population of some Asian countries.

#### Thorium content in skeleton

Th in skeleton was studied only in China, India and Republic of Korea. The highest content of 22.1  $\mu$ g Th was obtained in skeleton for adult population of Republic of Korea and the lowest content of 3.96  $\mu$ g for the Indian population. Th content of skeleton, even among the three countries, varied by a factor of more than five as can be clearly seen in Fig. 22. In view of the relatively low daily dietary intake of Th reported for the population of the Republic of Korea, such high skeleton content of Th appears surprising. The content of Th in skeleton of the Chinese population was 14.45  $\mu$ g. This high Th content for the Chinese population of course was consistent with its higher daily intake by the Chinese population. The ICRP-23 value for the Th in skeleton is 30  $\mu$ g [3].



FIG. 22. Contents of Th in skeleton in adult population of some Asian countries.

### Caesium in skeletal muscle

Cs contents of the skeletal muscle were estimated only in India, China and the Republic of Korea. The variation in the contents was less than a factor of 2. The values for the three countries ranged between 327  $\mu$ g for India to 624  $\mu$ g for Republic of Korea. The highest content of Cs in the skeletal muscle for the Republic of Korean population is clear from the box plots in Fig.23.

The Asian median skeletal muscle content was 469  $\mu$ g. It was lower than the 570  $\mu$ g value reported in ICRP-23. The lower Asian median could be explained on the basis of 20% lower body weight for Asian population [10] when compared with the Caucasian population represented by ICRP reference man. It was also marginally lower than 512  $\mu$ g reported for Japanese population by Nishiyama et al. [77]. The data on the dietary intake and the median organ content of Cs could be applied to make a theoretical derivation of the expected biological half-life of <sup>137</sup>Cs.



FIG. 23. Contents of Cs in the skeletal muscle in adult population of some Asian countries.

# Sr in skeleton

Sr is the stable counterpart of another important beta emitting fission product <sup>90</sup>Sr, which is released during operation of the nuclear power reactors. Contents of Sr in skeleton were reported from China, India and Republic of Korea. The contents of Sr ranged between 172.5 mg for the Indian and 412 mg for the population of Republic of Korea. The distribution of Sr in the skeleton of the populations of the three countries is well depicted in Fig. 24.

Tanaka *et al.* [78] reported 437 mg Sr as the skeleton content for the population of Japan, which compares well with the Asian median value of 353 mg (Table 39). A skeleton content of 320 mg Sr is reported in ICRP-23 [3]



FIG. 24. Contents of Sr in the skeleton in adult population of some Asian countries.

# 8.3. Special applications of data on elemental intakes and organ contents

Data on dietary intakes and organ contents of elements obtained in the CRP are useful for a number of applications in the field of radiological protection and nutrition. A few applications are discussed below.

# 8.3.1. Proposing daily dietary intake of some selected elements for the Asian population

Based on data obtained from Asian countries on intakes of elements of importance in radiological protection and also nutrition, the representative intakes (median) for the Asian population are proposed. These values (ranges and medians) are given in Table 35. Proposed data are listed along with data from ICRP-23, the global averages and the US RDA (for essential elements), for information. It may be borne in mind that ICRP reference man (also representing the majority of the US population) is about 20% heavier in weight as compared to the reference Asian Man [10]. Thus there is an extra requirement of calories (energy) for the population represented by ICRP reference man from consumption of diet, leading to enhanced daily intakes of trace elements.

As seen from the ranges of intakes presented in Table 35 there were inter-country variations in intakes of up to an order of magnitude for most of the elements. It was therefore considered reasonable to employ median values as representative intakes by the Asian population. For elements of importance in radiological protection, in relation to ICRP-23 data, the intakes are comparable for Cs and Sr and lower for elements Ca, I, K, Th and U. Among essential elements, intakes for Na, Mg, Mn, are comparable and for Cu, Fe, Zn and P lower. Intakes of Ca, I, Fe, Zn (essential elements) by the Asian population are also lower than the recommended dietary allowances proposed by the US National Academy of Sciences [79]. It may be interesting to observe that for most elements, the global averages

lie between intakes for the Asian and Caucasian populations. Another interesting feature of the comparison is that the data compared well with the global intake data proposed by UNSCEAR 2000 [68]. Reference intake values for both these elements may need downward revision compared with earlier data in the literature [3].

# 8.3.2. Proposed contents of Cs, I, Sr, Th and U in some selected organs for Asian population

Based on the organ content data obtained in four Asian countries, representative organ content values were proposed for the Asian population. It may be noticed from Table 41 that with the exception of U in kidneys and skeleton and Th in skeleton, the ICRP values, wherever available, were in good agreement with those reported from the Asian region.

# TABLE 41. A COMPARISON OF PROPOSED CONTENTS OF Cs, I, Sr, Th and U IN SELECTED ORGANS OF THE ASIAN POPULATION, WITH THE ICRP REFERENCE MAN VALUES

Organ	Element	Asian Value	ICRP-23 Value		
Organ	(unit of conc.)	Range	Median		
Thyroid	I (mg)	6.27–19.34 (3)	12.0	12.0	
Kidney	U (µg)	0.14–0.26 (4)	0.19	7.0	
Liver	Th(µg)	0.12–0.53 (5)	0.23	-	
	U (µg)	0.08–0.77 (5)	0.20	0.45	
Skeleton	Sr(mg)	172.5–412.0 (5)	353.1	320.0	
	Th(µg)	3.96–22.1 (4)	14.40	30.0	
	U (µg)	2.85–12.58 (4)	5.2	59.0	
Skeletal Muscle	Sr(mg)	327.0–623.6 (3)	469.0	570.0	
Lungs	Th(µg)	0.89–7.79 (5)	3.2	-	
	U (µg)	0.36–1.92 (5)	1.1	1.00	

The ICRP had not reported the organ contents for many of these important elements and therefore the data obtained in the present CRP could be considered as the first set of reliable data for their organ contents. The available ICRP data for the elements that were present at high concentrations in the organs are also shown in the table where the concentrations were low, limited or only questionable data were available. The U content of kidneys and skeleton are two such examples.

Consistently low contents of U in human kidneys (lower by an order of magnitude) and skeleton were obtained in all the Asian countries. The values are comparable to the contents reported for the New

York population of USA by Fisenne et al. [69]. Earlier literature data [3] indicating high values should be reviewed since such data are often used to predict certain biokinetic parameters of U and the unrealistically high values may affect the interpretation of its biokinetics and subsequently could lead to erroneous recommendations regarding limits on its annual intake (ALI).

# 8.3.3. Developing biokinetic parameters of trace elements and related radionuclides

As stated at the beginning of the Section, daily intakes of trace elements along with their organ contents could provide important information on some of their biokinetic parameters. Many workers have used such data to obtain biological half-life, gastrointestinal absorption factor and uptake of trace elements and their related radionuclides. Johnson and Lamothe used intake and organ content data to obtain gut-absorption factor for Th present in the diet [15]. Limson Zamora *et al.* have used similar data to obtain the gut-absorption factor of U in humans [18]. Dang *et al.* [16] used data on daily dietary intake and daily urinary excretion to obtain the gut-absorption factor of U for adult Indian population.

# 8.3.4. Estimation of the population specific biological half-lives for important radionuclides

The biological half-life  $(t_{b1/2})$  of a radionuclide or its stable isotope in the body could be derived by using the simple mathematical formula (assuming dynamic equilibrium between the amount of element or activity entering and leaving the body):

$$t_{b1/2} = \frac{0.693 \text{ MC}}{\text{I fi } f_2}$$

Where  $t_{b1/2}$  is the biological half-life (to be determined), M is the mass of the organ (source) which concentrates the stable element or the radionuclide (radioisotope) of interest. C is the concentration in the organ. MC together, therefore, represents content of the organ. I represents its daily intake.  $f_1$  denotes its fraction that is absorbed from the gastrointestinal tract and  $f_2$  represents the fraction of the absorbed quantity that goes to the organ. The product of  $I.f_1.f_2$  together, represents the quantity of the radionuclide entering the organ in a single day.

This formula is applicable in predicting the biological half-life when the excretion of a radionuclide from the body could be represented by a single exponential function. This is applicable to <sup>137</sup>Cs and <sup>131</sup>I, whose models are given in ICRP-30 and have not been changed by the ICRP during their current revision.

Nair et al. [80] used data generated during the current CRP (Indian contribution) on daily intake of Cs and its content in skeletal muscle to obtain the biological half-life of Cs for the urban Indian population. They predicted biological half-life of <sup>137</sup>Cs for the general Indian population comparable with that for ICRP reference man. But for the urban population, a relatively shorter half-life was predicted by them. The short half-life of <sup>137</sup>Cs for the urban population was reportedly due to higher daily intake of Cs through the consumption of relatively large quantity of milk products and vegetables by urban population when compared with the general population residing in rural areas of India.

# 8.3.5. Appraisal of some aspects of current ICRP Models using intakes and organ contents data

Both Cs and I are mainly distributed in soft tissues whereas Cs like K is uniformly distributed in soft tissue mass. I is retained mainly in thyroid, where it is utilized in the synthesis of thyroid hormone. Th, U and Sr on the other hand are mainly bone seekers, where they translocate from bone surface to bone volume and then to bone marrow and then get removed back into transfer compartment from these

three compartments very slowly, following much complex pathways in comparison to the Cs and I. Whereas, the biological half-lives of Cs and I could be directly obtained from data on their intakes and organ contents, in the case of Sr, Th and U, it is not possible to calculate their biological half-lives using the same method. These three elements follow more complex re-circulation pathways. Their data on intake and organ content however, could be employed to test their new ICRP models.

The new models for Sr, Th and U, along with those of Cs and I, were tested by calculating their organ contents by applying the new biokinetic models to the daily intakes obtained in the CRP for the population of China and India. The calculated organ contents were then compared with the actually measured organ content data. An agreement between the two values (within the limits of uncertainties) demonstrated the reliability of models. While calculating organ contents, the contribution from the inhalation route was assumed to be negligible and ingestion was considered to be the only source of the systemic organ content of trace elements. The calculations for the organ contents of these elements were carried out with the help of R.W. Leggett at Oakridge National Laboratory (ORNL), USA [88].

Only China and India were able to obtain truly representative data on intakes and organ contents. The Republic of Korea obtained only five sets of tissue samples for the organ content study, which were not sufficient to be statistically representative of the whole country. In the Philippines, organ contents were determined for liver and lungs only. Lungs, being part of the non-systemic compartment, reflect the input of trace elements mainly by inhalation and not by ingestion. Among the systemic organs, only 10 samples of liver were collected for the study and therefore the organ content data from the Philippines were also not adequate to employ for testing the new models.

The results of the application of the models to the respective intakes are therefore reported only for India and China. A comparison of the calculated and measured organ contents are shown in Table 42. While studying the comparison, the large biological variations from one subject to another in intakes and organ contents as well as the general nature of the ICRP models needs to be kept in mind. As may be seen from the Table 42, keeping in mind the possible variations in the intakes and organ contents, the calculated and observed values were within a factor of two. It may be interesting to note that when data from India and China are compared, the relatively higher elemental intakes by the Chinese population got reflected in their higher organ contents and calculated values also supported the data.

The comparison of calculated and measured organ contents for the trace elements have shown for the first time the consistency of data obtained at the environmental level of intakes, with new ICRP models thus suggesting their effective applicability to the internal dosimetry studies.

Another aspect of the data obtained in the CRP that supported the new model of Th, was liver to skeleton content ratio of Th. Data on Th in liver and skeleton for the Asian population were utilized to obtain the liver to skeleton content ratio of 0.03, which is consistent with the value 0.03–0.04 reported by Leggett on the basis of the current ICRP model for Th [81].

# 8.3.6. Estimation of internal dose due to the ingestion of environmental radioactivity

Annual committed effective radiation dose from dietary intakes of two naturally occurring long lived radionuclides <sup>232</sup>Th and <sup>238</sup>U could be measured using their specific activity data. Based on the data collected in the CRP on intakes of Th and U for the Asian population, the combined annual committed effective dose from the ingestion of <sup>232</sup>Th and <sup>238</sup>U to the Asian population was calculated to be 0.54  $\mu$ Sv, which is more than three orders of magnitude lower than the radiation dose of 1000  $\mu$ Sv recommended by ICRP-60 [82] for the general public.

# TABLE 42. A COMPARISON OF MEASURED AND CALCULATED ORGAN CONTENTS FOR ADULT ASIAN POPULATIONS

Element	Organ	Content		Daily intake
		measured	(*) calculated	
Thorium	Skeleton	3.50 µg	3.71 µg	0.64 µg
	Liver	0.14 µg	0.14 µg	
Uranium	Skeleton	2.90 µg	2.38 µg	0.54 μg
	Liver	0.07 µg	0.07 µg	
	Kidney	0.19 µg	0.04 µg	
Strontium	Skeleton	208 mg	369 mg	1.5 mg
Iodine	Thyroid	6.7 mg	4.95 mg	110.0 µg
Caesium	Skeletal muscle	298 µg	247 µg	4.35 μg

# India

# China

Element	Organ	Content		Daily intake	
		Measured	(*) calculated		
Thorium	Skeleton	19.7 µg	18.1 µg	3.13 µg	
	Liver	0.59 µg	0.70 µg		
Uranium	Skeleton	12.6 µg	22.2 µg	5.04 µg	
	Liver	0.77 μg	0.36 µg		
	Kidney	0.26 µg	0.70 μg		
Strontium	Skeleton	455 mg	708 mg	2.88 mg	
Iodine	Thyroid	17.6 mg	13.7 mg	327.0 μg	
Caesium	Skeletal muscle	620 µg	738 µg	13.0 µg	

(\*) Organ contents were calculated by applying the new ICRP models of the elements to their daily dietary intakes.

# 8.3.7. Strontium to calcium ratio in Asian diet

Another interesting observation in the comparative study of the daily elemental intakes by Asian and Caucasian population was much higher Sr/Ca intake ratio of  $3.7 \times 10^{-4}$  for Asians in comparison to  $1.7 \times 10^{-4}$  for Caucasians. Whereas Ca and Sr concentrations in plant are the direct reflection of their concentrations in soil, the animal gastrointestinal tract discriminates against Sr and therefore its absorption is lower than that of Ca. Hence the intakes ratio from the plant based diet is higher than the animal based diet such as dairy products. The Sr/Ca ratio in diet could therefore provide information about the source of their intakes. The relatively higher Sr/Ca ratio for the diet of Asian population could be attributed to consumption of plant materials (fruits, vegetables, cereals, etc.) as the source of the two elements. For Caucasians, most of the Ca and Sr are sourced from dairy products such as milk, cheese, etc. The intake of dairy products for Asian population is quite low as already shown in Table 36.

#### 8.3.8. Estimation of transfer factor between human body and diet by Sr/Ca ratio

When considering internal radiation dose due to radio strontium in the equilibrium state, Sr to Ca content ratio in bone can be related to that in the diet as its precursor: The observed Sr/Ca ratio between bone and diet (O.R.<sub>bone-diet</sub>), was used to estimate the extent of transfer of <sup>90</sup>Sr to human bone when appropriate biokinetic models for Sr were not available. It reflects the overall discrimination of strontium against calcium between sample and its precursor as originally proposed by Comar [83].

O. R.<sub>bone-diet</sub> = 
$$\frac{\text{Sr/Ca ratio in bone}}{\text{Sr/Ca ratio in diet}}$$
 (1)

As shown in Table 43, the average O.R.<sub>bone-diet</sub> for India, China and Japan was estimated as 0.11 in comparison to 0.19 calculated from the ICRP reference man data. The present value is also lower than data ranging between 0.16–0.25 for Europeans and North Americans, reported by Snyder [84] and UNSCEAR [85]. The result could be useful to assess uptake of <sup>90</sup>Sr in the Asian population when required to predict internal doses from it in an emergency situation.

# 9. CONCLUSIONS

The CRP has effectively contributed to strengthen the ability, in a group of Asian countries, to apply sensitive and robust analytical techniques (including sampling and sample preparation methods) for carrying out reliable analytical measurements of Cs, I, Sr, Th and U. The most significant outcome of this CRP was the generation of reliable data sets for dietary intake by each of the participating countries (and also for tissue samples in some countries) that will enhance their ability to resolve national problems of radiological protection, as well as facilitating the development of the characteristics of a reference Asian Man, the primary goal of this region. In addition, China, India and Japan have developed their own versions of a national reference man and Korea and Vietnam are well on the way to achieving the same objective.

# TABLE 43. Sr/Ca RATIOS IN ASIAN DIET AND BONE — ESTIMATION OF TRANSFER FACTOR: OBSERVED RATIO

			Countr	У								
	Bangladesh	China	India	Indo- nesia	Japan	Korea	Pak- istan	Philip- pines	Viet- nam	Mean	S.D	R.S.D
Sr intake, median (mg)	1.83	2.88	1.5	2.2	1.60	1.64	2.43	1.36	1.29			
Ca intake, median (g)	0.31	0.68	0.36	0.3	0.44	0.43	0.48	0.49	0.64			
Sr/Ca ratio, mean $(\times 10^{-3})$	6.2	4.0	4.3	8.0	3.4	4.1	5.8	5.2	1.9	4.4	1.4	31.8
Sr/Ca, median (× $10^{-3}$ )	5.1	4.2	4.2	7.3	3.6	3.8	5.5	2.8	2.0	3.9	1.1	28.2
Sr in bone, mean (mg)		457.4	235.7	850	437	436.7						
Ca in bone, measured/ estimated (g)		987.5	680	972	850							
Sr/Ca in bone ( $\times 10^{-3}$ )		0.457	0.347	0.875	0.51	0.514				0.438	0.083	18.9
Sr/Ca O.R. (bone-diet) o intake	on median	0.11	0.083	0.12	0.14					0.11	0.030	27.2
Japanese in 1960s and 1	970s (ref. 1)									0.12		
Western Europeans and Americans in 1960s (ref	North f 2, 3)									0.16-0.25		

Table references:

1) TANAKA, G. et al.: Health Phys. 40, 601–614, 1981 [78]

2) SNYDER, W. S. et al.: Health Phys. 10, 171-182, 1964 [84]

3) UNITED NATIONS: A/7613, 1969 [85].

Harmonized protocols developed during the CRP for collecting and processing diet and tissue samples were followed by all the participating countries. The CRL established at the NIRS, Japan, took responsibility for internal and external quality control. As a result of these efforts, the general level of competence in analytical quality assurance has been raised in the participating Asian countries. More specifically, improved reference values have been derived for a number of elements and RMs of interest. In addition, the CRP participants made significant contributions to the development and certification of a new RM (Typical Japanese Diet) for use during the CRP.

Results of the analysis of diet samples brought out clearly two facts: (i) very large inter- and intracountry variations of up to an order of magnitude in the intakes of trace elements; (ii) lower median intakes for all the elements of importance in radiological protection in Asian populations in comparison with the intake values reported in ICRP-23.

With the exception of Mn and Mg, the mean intakes of essential elements in Asian countries were also lower than the intakes reported in ICRP-23 [3]; they were also lower than the daily dietary allowances

recommended by the US National Academy of Sciences [79]. However they were not much lower than the global median values reported by the IAEA [10].

For individual countries, the intakes of Fe in China, Pakistan and India appeared to be adequate while in Japan, Philippines and Vietnam they were low in comparison both with global intake values [10] as well as with the US RDAs [79]. However, these observations do not take account of possible differences in bioavailability and, therefore, its impact on sufficiency or deficiency status of a particular element in a given country. In this connection there may be a need to re-examine the current dietary allowances of various elements in Asian populations, especially in view of the consistently lower intake values of Ca, Zn (and of Fe in Japan; a developed country in the Asian region), also taking account of bioavailability as a factor.

In the light of the report in ICRP-67 that extremely low Ca intakes may influence the gastrointestinal absorption of <sup>90</sup>Sr, one of the abundant fission products, the significantly low intakes of Ca in the Asian region may need to be reviewed.

Data for human organ contents of Cs, I, Sr, Th and U, were collected only for those organs that tend to concentrate these elements and therefore their related radionuclides. I was measured in thyroid, Cs in skeletal muscle, Th in skeleton, Th and U in lung and liver, U in skeleton, lung, kidney and liver and Sr only in the skeleton. Some participants also generated additional data on these and some other trace elements in as many organs as they could analyse. Bangladesh, Pakistan, Indonesia and Vietnam did not obtain any organ content data for trace elements due to cultural problems which hindered the collection of suitable samples.

The organ content data generated in this CRP were consistently lower than the values reported in ICRP-23 and ICRP-30. For U the median Asian kidney content of 0.19  $\mu$ g was nearly 40 times lower than the ICRP-23 value of 7.0  $\mu$ g. The implications of this observation in connection with the biokinetic of U deserve to be studied in the near future.

Data on the intakes of Cs, I, Sr, Th and U were applied to the new ICRP models. The calculated organ contents compared well with those measured in the CRP, indicating the consistency of the organ content data generated in the CRP with the new ICRP models. The ratio of the Th content in liver to that in bone observed in some of the countries agreed well with the new model for Th. Such consistency in the data generated at environmental levels of exposure provides credence and reliability to the new ICRP models in comparison with the old models described in ICRP-30.

The data on organ contents generated by the CRP were utilized by some participants to predict biokinetic parameters such as the biological half-lives of I and Cs and their related radionuclides <sup>137</sup>Cs and <sup>131</sup>I. The calculated half-lives were found to be only marginally different from those proposed by ICRP.

The CRP had also been beneficial to the participating countries in terms of the exchange of experts and scientific workers between one country and another. This has enabled sharing knowledge and expertise not only in the field of analytical methodology but also in the field of radiological protection. The enhanced analytical capabilities of the participating countries led to increased research efforts. The work carried out in this CRP has so far resulted in more than 20 scientific peer-reviewed publications by the participants.

The organ data generated in non-Muslim neighbouring countries in Asia with similar geographical location, eating habits and lifestyle could be used meaningfully by Muslim countries for the purpose of realistic radiation dosimetry as organ samples from the Muslim countries participating in this CRP were not available for the biokinetic study.

# 10. FUTURE NEEDS

Although this CRP has already been significant in facilitating efforts to use national models as the basis for a reference Asian Man, there is still a need to obtain additional data in the Asian region on the following topics:

- Intake studies in infants and children (0, 5, 10 and 15 a) for the same elements of importance in radiological protection as are covered in this CRP.
- Excretion studies for the same elements to define biokinetic parameters such as the gastrointestinal absorption factor,  $f_1$ , to be used together with the already obtained intakes and organ contents data.
- Physiological data for adults and children, e.g. respiratory dynamics and water balance.
- Information on airborne particulate matter in Asian countries, particularly with respect to the elements of radiological importance. This could be useful to complete the picture on the biokinetics of the elements of radiological importance studied in this CRP.
- Biokinetic data (e.g. the biological half-life) for <sup>131</sup>I, <sup>137</sup>Cs, <sup>3</sup>H, etc., such as can be studied in accidentally over-exposed persons or patients undergoing nuclear medicine treatment.

In view of concerns over the increased incidence of thyroid cancer in some exposed children following the Chernobyl nuclear power plant accident, greater attention is now being given to preventing the deposition of radioiodine in the thyroid of children, especially infants, as one of the critical activities of emergency preparedness. Administration of potassium iodide as a thyroid blocker can cause side effects. Therefore, a knowledge of the daily iodine intake in young children will be beneficial to preestimate doses to them in an emergency situation and also to monitor the effectiveness of stable iodine administration. As intake and excretion levels of stable iodine may vary from one country or region to another, studies on metabolic data for iodine in younger children may be of urgent interest in Asian regions where more nuclear power reactors are expected to be built.

# 11. ADDENDUM: RELEVANCE OF ICRP PUBLICATION 89 TO THIS REPORT

Some time after the Steering Committee had finished drafting this report, the Committee became aware of a new ICRP publication, No. 89 [89], which appears to overlap somewhat with the subject matter of this IAEA report. Mainly, however, it is of relevance to the earlier IAEA report describing phase 1 of the reference Asian Man Project [10].

# ICRP Publication 89 and Reference Asian Man Project, Phase 1

ICRP-89 was published in 2003 to "revise and extend the information in Publication 23 as appropriate, and to provide additional information on individual variation among grossly normal individuals resulting from differences in age, gender, race, or other factors". In this Publication, reference values for further anatomical and physiological data for adult males and females and children are presented.

Results of the IAEA project (phase 1), particularly height, weight and mass of internal organs, are extensively quoted in ICRP-89 in drawing comparisons between European and Asian data (see pages 83–85, 101–102, 121–122, 124–125, 135–136, 147–148, 206–209, and 222–225 of reference [89]). It is relevant to note here that phase 1 of the IAEA-RCA Reference Asian Man Project had previously been endorsed by ICRP Committee 2.

# **Reference values on elements in ICRP Publication 89**

ICRP-89 also gives reference values on the composition of body tissues for children and adults for 13 major and minor elements "for use in evaluating the transport of radiation within the body and its energy deposition in the various tissues" (see pages 243–246 and tables 13.1–13.6 of reference [89]). Data on eleven elements are also given for the newborn. These elements include H, C, N, O, Na, P, S, Cl and K in various soft tissues, Ca in the skeleton and kidney, Fe in the blood and I in the thyroid. No data for other elements in body tissues, and no data for the ingestion intake were included as physiological data.

The current IAEA project studied the ingestion and organ content of Cs, Sr, I, Th and U, and K and Ca, chemically similar elements to Cs and Sr, respectively, that are important in radionuclide metabolism in man, and that had been selected in consultation with the ICRP Reference Man Task Group. The IAEA project makes no attempt to propose reference values for K in the muscle or Ca in the skeleton. Iodine in the thyroid appears to be the only commonality between ICRP-89 and the present report.

# Comparison of thyroidal iodine between ICRP Publications and the present report

As shown in Table 39 of this document, the Asian median of thyroidal iodine content is 12 mg, which is identical to the figure proposed by ICRP in ICRP-23 (page 303 of reference [3]).

The *newer* ICRP value given in ICRP-89 is quoted only as 0.1 % by mass. For comparison, the new Asian value proposed in this report (12 mg in the total organ) translates into a value of 0.05% by mass. This is on the basis of an organ weight of 23.2 g (see page 45 of reference [10]).

Comparison of these two values (0.1% by mass in ICRP-89 and 0.05% by mass in the present report) is very imprecise given the fact that the ICRP-89 value is only quoted to one digit of significance. Both sets of figures probably also have wide confidence intervals (although these are nowhere stated). The only conclusion that can presently be drawn is that both sets of values appear to be compatible with each other.

# **ABBREVIATIONS**

AAS	Atomic Absorption Spectrometry
ALI	Annual Limit of Intake
BSS	Basic Safety Standards
CRL	Central Reference Laboratory
CRM	Certified Reference Material
DAC	Derived Air Concentration
ENAA	Epithermal Neutron Activation Analysis
$\mathbf{f}_1$	Gastrointestinal absorption factor
$\mathbf{f}_2$	Organ uptake factor
ICP-AES	Inductively Coupled Plasma Atomic Emission Spectrometry
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICRP	International Commission on Radiological Protection
INAA	Instrumental Neutron Activation Analysis
MCA	Multi Channel Analyser
NIES	National Institute for Environmental Studies (Japan)
NIRS	National Institute of Radiological Sciences (Japan)
NIST	National Institute of Standards and Technology (USA)
QA/QC	Quality Assurance/Quality Control
RCA	Regional Cooperative Agreement for Research, Development and Training Related to Nuclear Science and Technology for Asia and the Pacific
RDA	Recommended Dietary Allowance (US)
RM	Reference Material
RNAA	Radiochemical Neutron Activation Analysis
SRM	Standard Reference Material (NIST)
$t_{b1/2}$	Biological half-life
WHO	World Health Organization
•	•

1 nCi = 37 mBq 1  $\mu g^{238}U = 12.4$  mBq 1  $\mu g^{232}Th = 4.1$  mBq.

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### Publications describing the project as a whole:

IYENGAR, G.V., KAWAMURA, H., PARR, R.M., MIAH, F.K., WANG, J., DANG H.S., DJOJOSUBROTO, H., CHO, S.Y., AKHTER, P., NATERA, E.S., MONG N.S., Dietary intake of essential minor and trace elements from Asian diets, Food and Nutrition Bulletin **23**(3) (2002) 124–128.

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# IAEA MEETINGS RELATED TO THIS PUBLICATION

# **Project Formulation Meeting**

Hitachinaka, Japan, 27 February-3 March 1995

# **Research Coordination Meetings**

Manila, Philippines, 1–4 July 1996 Taiyuan, China, 15–19 June 1998 Dalat, Vietnam, 26–30 June 2000

# **Consultants Meetings\***

Vienna, Austria, 22–24 April 1996 Taiyuan, China, 14 June 1998 Kona, Hawaii, USA, 6–7 April 2000 Vienna, Austria, 25 February–1 March 2002 Vienna, Austria, 24 to 28 June 2002 Vienna, Austria, 19–23 May 2003

\* Meetings of the Steering Committee

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