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Health and environmental aspects of nuclear fuel cycle facilities



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HEALTH AND ENVIRONMENTAL ASPECTS OF NUCLEAR FUEL CYCLE FACILITIES

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FOREWORD

Energy is essential for human life. Urbanization, industrialization and a rising standard of living will lead to growing energy demand. This is a major factor which will give rise to increasing use of energy in the form of electricity. In some highly industrialized countries the relative rate of growth in energy demand is slowing down, and in some countries the demand may even be decreasing, but this is more than offset by increased rates of growth in the developing countries. At the same time, health and environmental concerns are high on the agenda of decision makers and the public, as shown by the United Nations Conference on Environment and Development (1992), the World Energy Council (1992) and the Helsinki Symposium (1991).

Despite conservation and increased efficiency in the use of energy, a variety of economically available energy sources will have to be used to meet future energy demands. The selection of appropriate energy sources is a policy matter for national authorities and involves economic, environmental and technical issues, including availability of national resources and balance of payments. The environmental constraints and advantages of the various energy sources need to be objectively analysed. The use of any energy source has some effect on the environment. It is difficult to compare the impacts on environment and public health of all energy sources, but such comparisons are necessary if well informed decisions are to be made.

To improve the ability for comparative assessment of energy chains for electricity generation, IAEA has been carrying out a project Comparative Assessment of the Health and Environmental Impact of Nuclear Power and Other Energy Systems as part of Subprogramme X.03: Comparative Assessment of Nuclear Power and Other Energy Sources, in co-operation with other international organizations.

Part of this programme is the DECADES (Database and Methodologies for Comparative Assessment of Different Energy Sources for Electricity Generation) project. DECADES is an inter-agency joint project to establish the databases and methods needed to carry out comparative assessment of different energy sources for electricity generation. The objective of the DECADES project is to enhance the capabilities of incorporating health and environmental issues in the planning and decision making of the electricity sector.

The purpose of the present publication is to give a generic description of health and environmental aspects of nuclear fuel cycle facilities. Primarily the report is meant to stand alone; however, because of the content of the publication and in the context of the DECADES project, it may serve as a means of introducing specialists in other fuel cycles to the nuclear fuel cycle.

The report consists of three parts. Part I reviews health and environmental aspects of nuclear fuel cycle facilities. Part II contains site reports and Part III includes country reports which were presented at a series of Technical Committee Meetings held between December 1992 and November 1994 on the Health and Environmental Impacts of Nuclear Fuel Cycle Facilities.

The present work began in 1992 and involved five consultants meetings and three Technical Committee meetings. The IAEA wishes to express its gratitude to all those who participated in the preparation of this publication and also to the Member States that sent experts to assist the IAEA in this work. Special thanks are due to R. J. Maloney, Atomic Energy Control Board, Canada, who edited the text for technical content and prepared it for publication.

EDITORIAL NOTE

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PART I

**OVERVIEW OF HEALTH AND ENVIRONMENTAL ASPECTS
OF NUCLEAR FUEL CYCLE FACILITIES**

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1. INTRODUCTION

1.1. PURPOSE AND SCOPE

This publication is primarily intended to stand alone; however in the context of the DECADES project it may also serve as a means of introducing specialists in other fuel cycles to the nuclear fuel cycle. In order to carry out a fair assessment of the different energy chains, it is important that specialists doing similar work in other energy chains have a general knowledge of all parts of the nuclear fuel cycle. Thus this TECDOC can be considered as an adjunct to the DECADES programme. Although some data are presented here, the reader is referred to the appropriate parts of the DECADES database for more detailed information.

A general approach has been adopted because the purpose of the document is to give a generic description of the environmental releases from the nuclear fuel cycle and health effects on workers in the industry. In keeping with this general approach the document discusses only the releases from nuclear facilities since impacts from these releases are considered to be highly site and country specific. For example the definition of environment may differ from country to country, as may the end points used to define impacts. Thus in only a few cases and for illustration purposes are the impacts of these releases discussed. The releases form the source terms and hence the starting point for impact assessments using site and country specific parameters.

It is the purpose of this publication to identify the main contaminants in the waste streams and effluents arising from nuclear facilities. In order to provide balance to the description of the releases from nuclear facilities, both radiological and non-radiological substances are included in the inventories of the releases where such data are available.

In carrying out the comparative assessment of different energy chains, it is important that impacts from all parts of the fuel cycle be considered. This would include the extraction of the raw material, transportation of intermediate products and wastes, processing materials, storage of used fuel or reprocessing of used fuel, and finally the management of the wastes generated in all steps of the chain.

The present report gives a structured presentation of health and environmental aspects of nuclear fuel cycle facilities. For each step of the nuclear fuel cycle, the basic process is first presented, then resource requirements and releases are estimated per annual GW(e) generation basis. Potential health and environmental impacts during routine operation are assessed radiologically and non-radiologically to the workers and the environment.

The report is limited to chosen health and environmental aspects of the nuclear fuel cycle facilities, namely to evaluation of collective and in some cases individual average doses and radioactive releases from the facilities. It also does not consider non-radiological hazards to the workers and the public, so that non-radiological health effects connected with the construction, operation and decommissioning of facilities are not discussed.

The reader looking for guidance as to the structure of a comprehensive health and environmental impact analysis, which should cover all stages of the nuclear fuel cycle and all kinds of health and environmental impacts, is referred to other IAEA documents.

1.2. NUCLEAR FUEL CYCLE DESCRIPTION

The main activities in the nuclear fuel cycle, comprise the following: mining, milling, refining and conversion, enrichment, fuel fabrication, spent fuel reprocessing, waste treatment and disposal. The scope of this report does not include nuclear power plants. There are two possible configurations of the fuel cycle:

- (a) The once-through fuel cycle, in which the ore is made into fuel passed through the reactor once and is then stored in waiting for final disposal (Fig. 2).
- (b) The reprocessing cycle, in which the fuel is passed through the reactor, reprocessed and passed through the reactor again (Fig. 3).

The difference between the once through cycle and the reprocessing cycle is that the reprocessing cycle makes more efficient use of the fuel through extraction of Pu and recycling of ^{235}U . This recycling can decrease the demand for natural uranium in these reactors by up to 35%.

The supporting manufacturing operations to the nuclear fuel cycle and construction of facilities have been judged to be relatively remote, indirect and less significant contributors to the environmental impact. On the other hand, decommissioning of nuclear facilities is important due to the treatment and disposal of contaminated materials and the amount of waste. The present report includes a brief description of decommissioning of facilities [1].

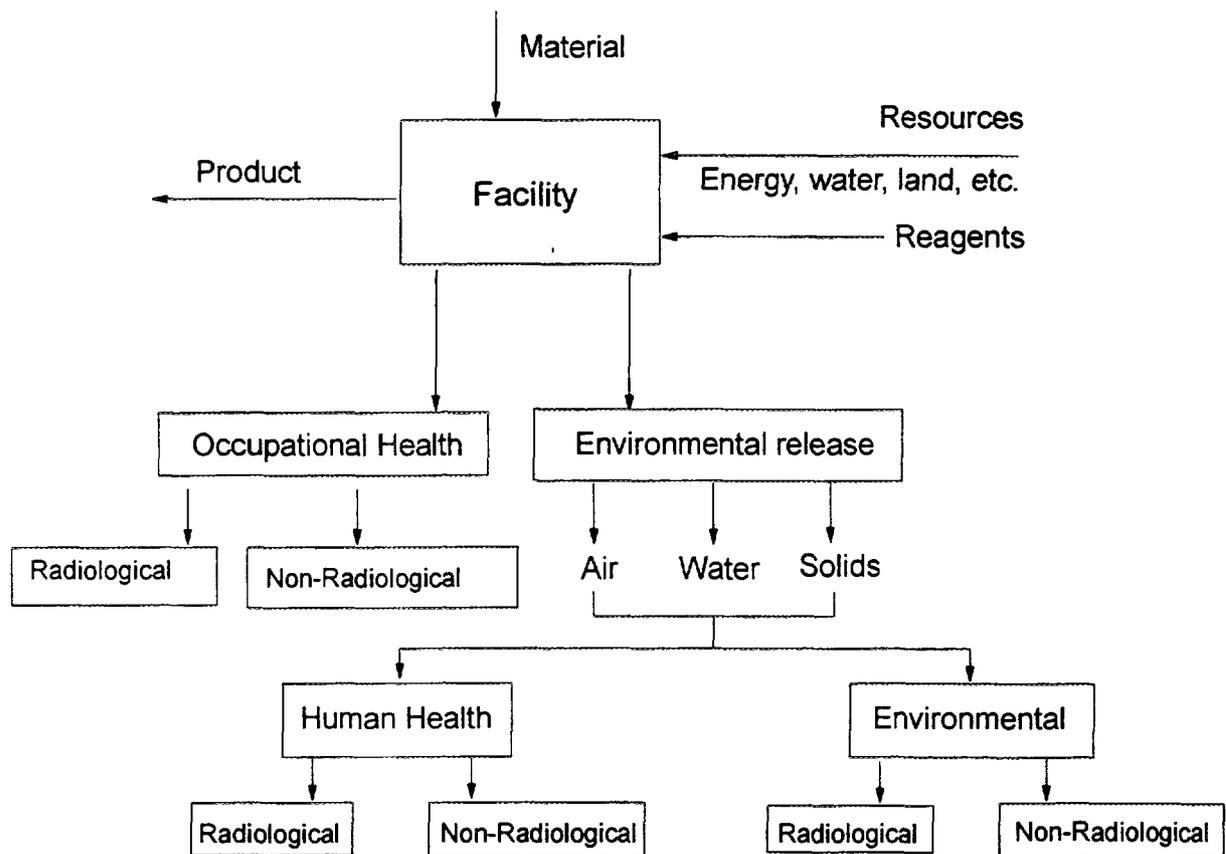


FIG. 1. Health Impacts - occupational and public

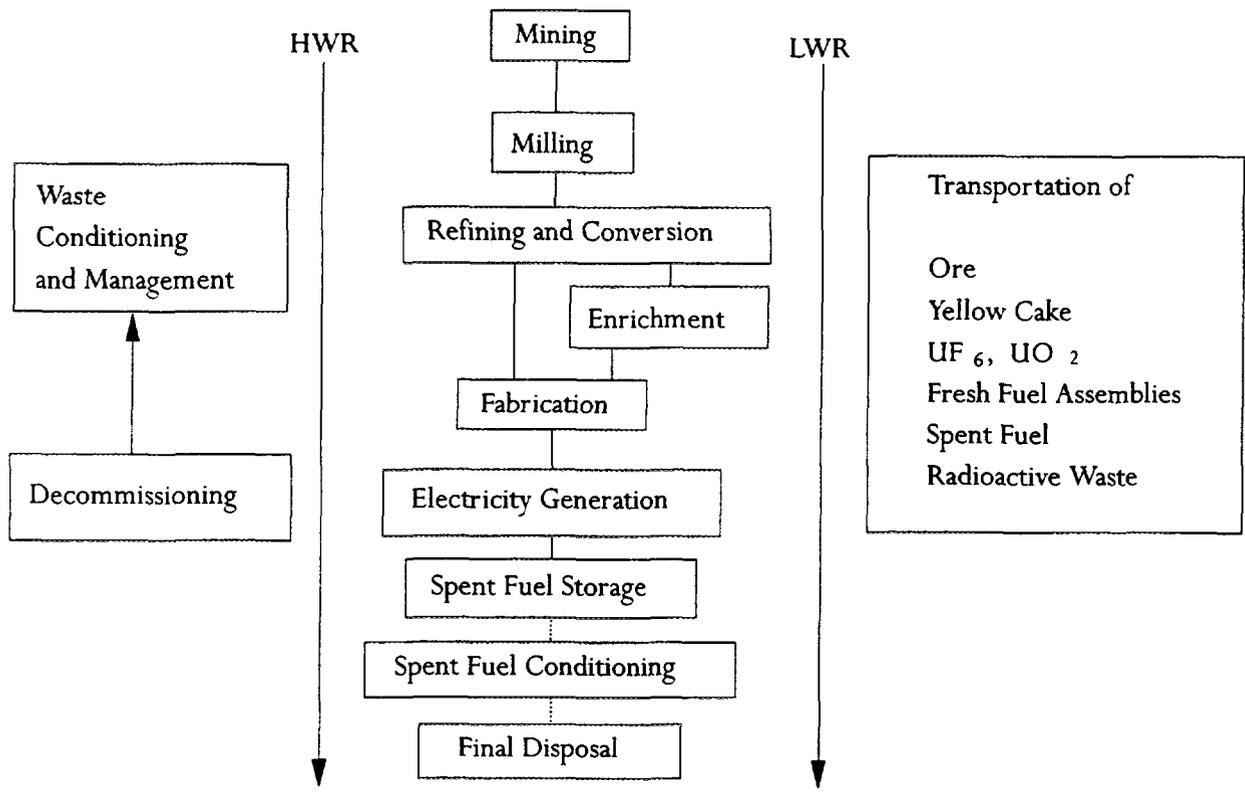


FIG. 2. Once-through of LWR & HWR

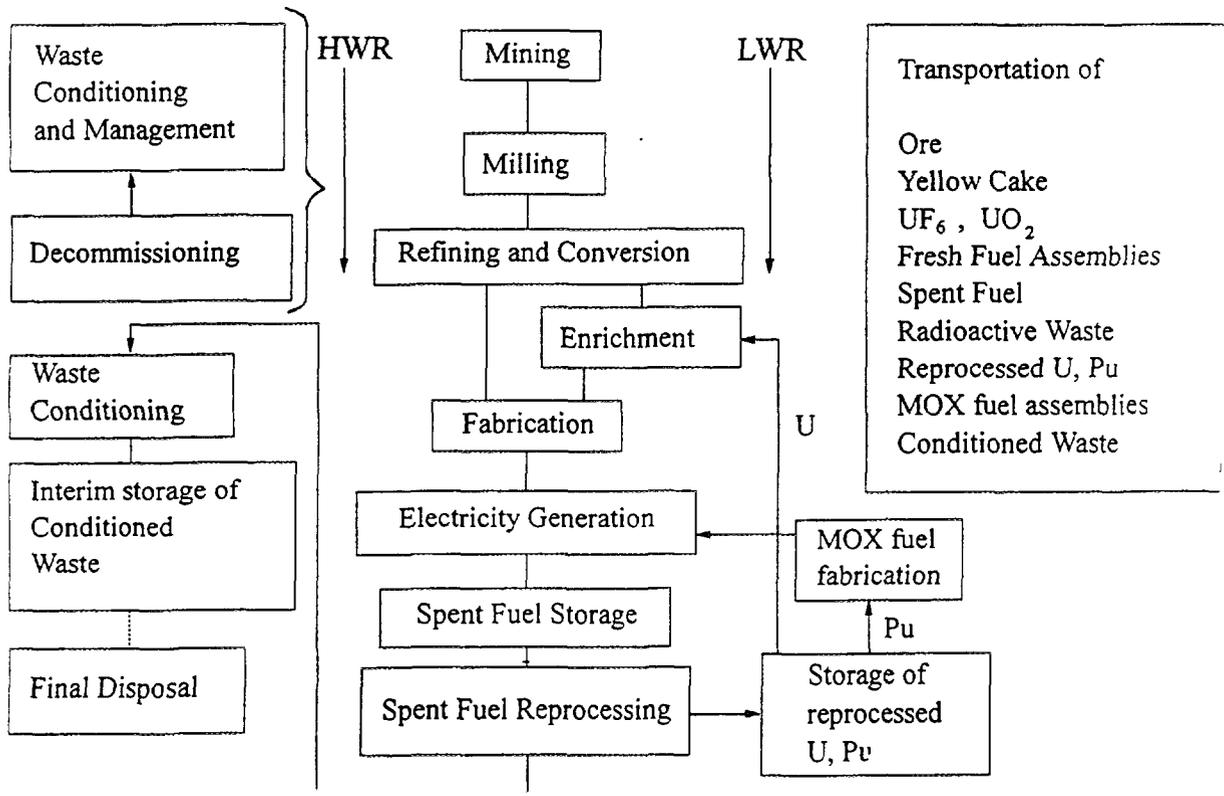


FIG. 3. Reprocessing cycle of LWR & HWR

2. POTENTIAL HUMAN HEALTH AND ENVIRONMENTAL IMPACTS

2.1. ENVIRONMENTAL IMPACT ASSESSMENT

In this document the environment is considered to be composed of interacting systems which in turn comprises biological elements, fauna and flora, and physical elements, atmosphere, land and water. Impacts are considered to be those effects that alter the existing system either temporarily or permanently. However the definition and measurement of impact is highly dependent on location, country, and social and economic factors.

Fig. 1. illustrates the flow of materials and wastes in a generic facility. Each nuclear fuel cycle facility produces a product from raw materials by using resources (energy, water, land) and reagents producing solid wastes and releasing effluent to air, water, and the soil. These radiological and non-radiological releases may have impacts on both humans (workers and the general public) and non-humans, and on the physical environment as in changes in water, sediment or air quality. Because of the long lived nature of some of the radionuclides released, concentration and biomagnification in all food chains must be taken into account where appropriate when evaluating environmental impacts.

Increasingly environmental assessment is becoming mandatory in many countries as part of the regulatory practice. Some reports have already been published on the effect of nuclear fuel cycle on the environment such as "Environmental Survey of the Uranium Fuel Cycle" (WASH-124B, U.S. AEC, 1974) [2]. It assesses the environmental considerations related to the nuclear fuel cycle for LWR. The environmental considerations included natural resource use (land, water, and fossil fuel), effluent (chemical, radiological and thermal) and those effects on the environment. "The Environmental Impacts of Production and Use of Energy, Part II, Nuclear Energy" (UNEP, 1979) [3] was published as part of the environmental impacts of different sources of energy. It deals with non-radiological and radiological impacts of each step of nuclear energy use. "Nuclear Energy and the Environment" (UNEP, 1980) [4] is also an outcome of the same project. "Nuclear Power, the Environment and Man" (IAEA, 1982) [5] is intended to present technical and other information on nuclear power and its health and environmental influences.

Information on radiological impact analysis may be obtained from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) report 1993 [6].

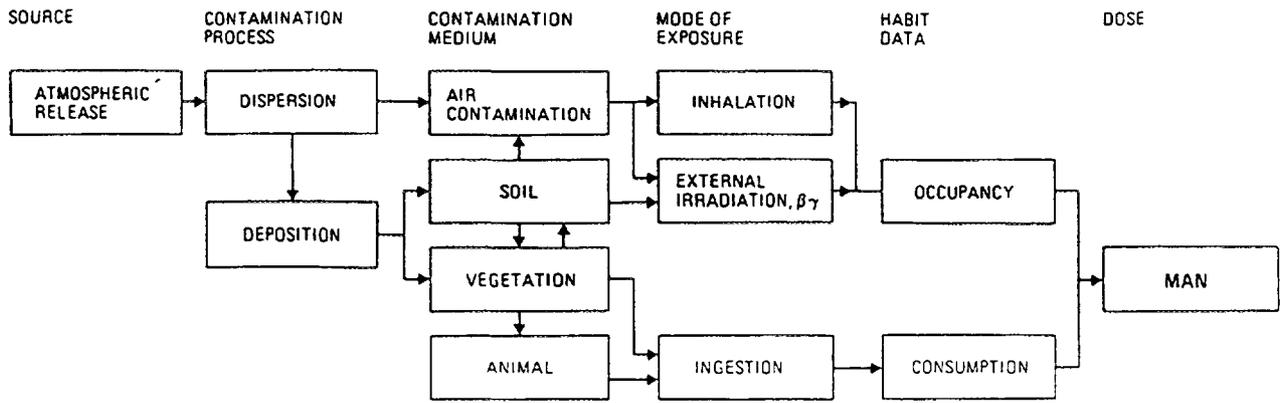
Within the nuclear fuel cycle the concept of nuclear safety analysis can also be used in environmental assessment in addressing accident conditions. Nuclear safety analysis is a means of analysing and evaluating the probability of accident occurrences, and identifying the measures that are necessary to reduce the probability and consequences of accidents. The release of hazardous materials is minimized by appropriate countermeasures, and health and environmental impacts are reduced. The OECD/NEA published a review of the safety of the nuclear fuel cycle facilities in 1993 [7]. The present report focuses on the normal operation of nuclear fuel cycle facilities and briefly refers to accident considerations.

2.2. RADIOLOGICAL IMPACTS

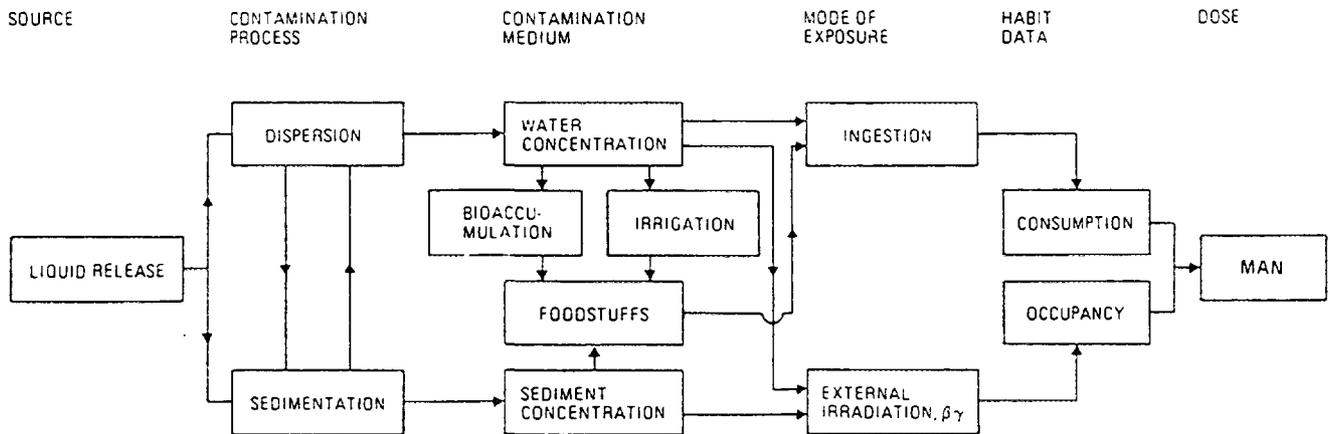
During normal operation small amounts of radioactive materials may be released into the environment resulting in some radiological impact on the general public and the environment.

2.2.1. Impacts on humans

Impacts on humans in the nuclear fuel cycle are stated in terms of dose. Doses to the public may be calculated using a method known as pathways analysis. Fig. 4 shows a typical set of pathways used in doing this calculation. In order to perform this calculation a knowledge of the transfer pathways in the human food chain and the transfer parameters between each compartment in that food chain is required.



Schematic representation of atmospheric pathways.



Schematic representation of aquatic pathways.

FIG. 4. *Typical set of pathways for analysis*

Occupational doses to workers on the other hand are usually assessed using direct measurements rather than modelling. International organizations such as UNSCEAR has done much to systematically collect, analyse, and present information in this area.

In its 1993 report to the General Assembly, the Committee evaluated the normalized collective doses to the public and annual occupational exposures to monitored workers from the various steps of the nuclear fuel cycle (Table I and II). The local and regional normalized collective effective doses, which are effectively received within one or two years of discharge, amount to 3 man Sv per GW(e).a and are principally due to routine atmospheric releases during reactor and mining operations.

The annual effective doses range from 0.001 - 0.02 mSv to the most highly exposed members of the public for the principal types of power plants. The corresponding annual figures for modern fuel reprocessing plants are 0.01 - 0.05 mSv.

The impact of the nuclear fuel is also quantified by using the concept of collective dose. This allows consideration of the impact on a local or regional level and on a global level. Data from UNSCEAR [6] showing the collective dose resulting from different parts of the nuclear fuel cycle are presented in Tables I and II for illustration. Details of the calculation of these data can be found in UNSCEAR [6].

Table I. Normalized collective effective dose to the public from the nuclear fuel cycle [6]

Source	Normalized collective effective dose (manSv per GW(e).a)
Local and regional component	
Mining	1.1
Milling	0.05
Mine & mill tailing (releases over 5 years)	0.3
Fuel fabrication	0.003
Reactor operation	1.34
Reprocessing	0.25
Transportation	0.1
Total (rounded)	3

Table II. Occupational exposures to monitored workers [6]

Occupational category	Annual collective effective dose (manSv)	Annual effective dose per monitored worker (mSv)	Normalized collective effective dose (manSv per GW(e).a)
Mining	1200	4.4	4.3
Milling	120	6.3	0.44
Enrichment	0.4	0.08	0.02
Fuel fabrication	22	0.8	0.07
Reactor operation	1100	2.5	4.3 (PWR), 7.9 (BWR)
Reprocessing	36 (LWR:5.7)	3.0 (LWR:1.4)	0.65
Research	100	0.8	1
Total (rounded)	2600	2.9	11 (PWR), 15 (BWR)

The global collective dose for the nuclear fuel cycle has been calculated to be about 200 man Sv (GW(e).a)⁻¹ [6]. This collective dose represents the total radiological impact on the population of the earth resulting from the nuclear fuel cycle. As a reference, the typical annual effective dose of 2.4 mSv from natural sources results in an annual collective dose to the world population of 5.3 billion people of about 13 million manSv. [6] (Fig.5).

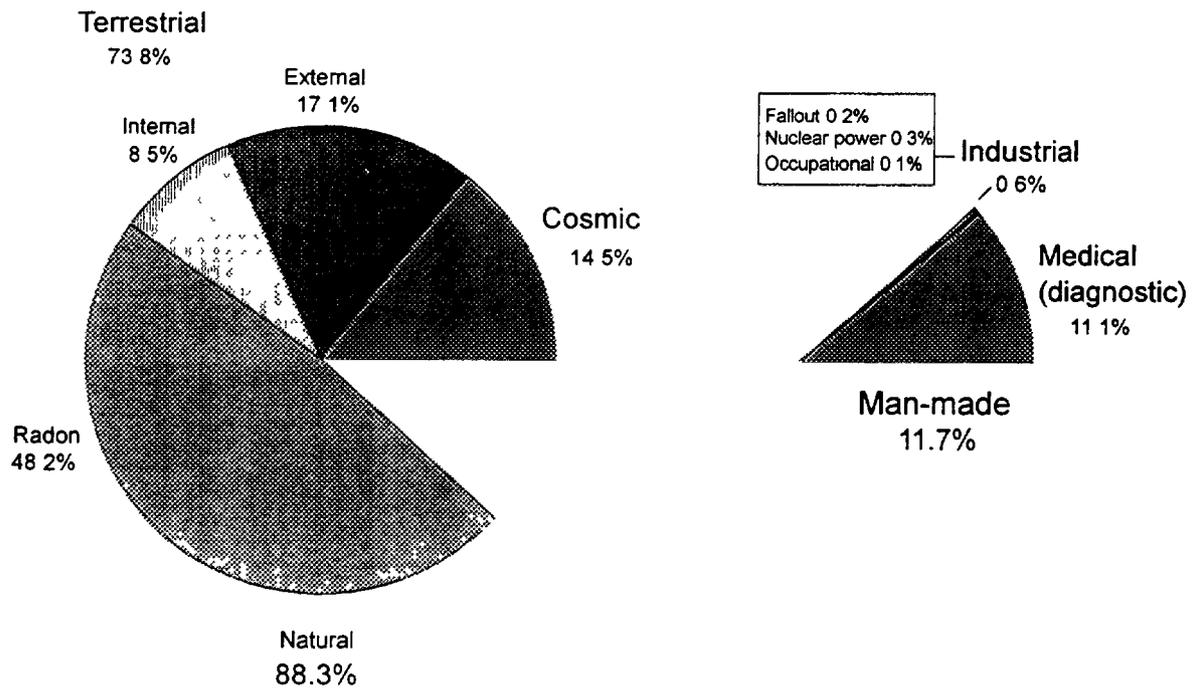


FIG. 5. Estimates of global average annual doses based on UNSCEAR 1993

2.2.2. Impacts on the environment

The general public is showing an increased concern with environmental protection and with the protection of species other than humans. In response to this increased concern, the IAEA, proposed that environmental impact comprise three elements [8]:

- a) Impacts on humans
- b) Impacts on resources (i.e. agricultural land, potable water etc)
- c) Impacts on the ecology

Concerning the protection of fauna and flora, recent analyses based on generalized information and conservative assumptions, have shown that radiation levels implied by current radiation protection standards for the public, are generally adequate to protect the other species (plants and animals) and that only the combination of specific ecological conditions such as the presence of rare or endangered species and specific stresses may require site specific analyses [9].

The environmental protection strategy for nuclear fuel cycle facilities is aimed at achieving national and international radiation standards for humans. As part of this strategy environmental monitoring and studies are carried out during all phases of the facility's operation.

2.3. NON-RADIOLOGICAL IMPACTS

2.3.1. Impacts on humans and environment

Many different chemicals and equipment are used in nuclear fuel cycle facilities. They are the same in many instances as used in other industries. Thus the effects of using these chemicals and equipment are also much the same. Mitigative measures consist of proper codes of practice, appropriate treatment systems, and a good safety culture.

Similarly, the environmental protection strategy for non-radiological substances released to the environment by nuclear fuel cycle facilities will be the same as other non-nuclear industries. Environmental monitoring and studies carried out for radiological contaminants will also include the monitoring and studies of non-radiological contaminants. Table IX shows potential sources of environmental impacts and mitigation.

Table III. Normalized release and collective doses from milling [6]^a

Radio-nuclide	Normalized release (GBq(GW(e).a) ⁻¹)			Normalized collective effective dose (man Sv (GW(e).a) ⁻¹)		
	Mill	Mill tailings		Mill	Mill tailings	
		In operation	Abandoned		In operation	Abandoned
Pb-210	0.02	20 000	1 000 ^b	0.00002	0.3 ^c	150 ^d
Po-210	0.02			0.00002		
Rn-222	3 000			0.045		
Ra-226	0.02			0.00001		
Th-230	0.02			0.0006		
U-234	0.4			0.003		
U-238	0.4			0.003		
Total			0.05			

- a. Normalized emissions in liquid effluents (0.01 for ²¹⁰Pb and ²³⁰Th; 0.02 for ²²⁶Ra; 0.3 for ²³⁴U and ²³⁸U) contribute negligibly to the collective dose.
- b. Annual activity released; the rate of activity is assumed to remain constant over more than 10,000 years.
- c. Dose commitment corresponding to a five-year release.
- d. Dose commitment corresponding to a 10,000 year release.

Table IV. Typical isotopic composition (plutonium originating from LWR Spent Fuel with 35 GW day/t burnups)

Pu 238	1.5 %
Pu 239	58.6 %
Pu 240	23.8 %
Pu 241	11 %
Pu 242	4.8 %
Am 241	1 - 3 %

Table V. Atmospheric emissions in MOX fuel fabrication

Annual atmospheric emissions (Bq/a)	10 E + 6 - 10 E + 7
Normalized annual emissions (Bq/GW(e).a)	10 E + 6 - 10 E + 7

Table VI. Radiation exposure in MOX fuel fabrication

	Existing Plants (experience)	Modern Plants (design)
Annual effective dose per monitored worker (Sv/a) (average)	0.007	0.003
Collective effective dose per produced MOX ton (man Sv/ton)	0.07	0.005 - 0.020
Normalized collective effective dose (man Sv/GW(e).a)	2.0	0.15 - 0.60

Note: To date, MOX fabrication world capacities are about 100 t/y; additional plants with capacities of about 400 t/y are expected to be commissioned before 2000.

Table VII. Occupational exposure from fuel reprocessing [6]

Oxide fuel	Monitored workers (thousands)	Annual collective effective dose (manSv)	Annual effective dose per monitored worker (mSv)
1975-1979	0.1	0.36	4.0
1980-1984	1.0	2.4	2.3
1985-1989	4.0	5.7	1.4

Table VIII. Typical characteristics of waste classes [17]

Waste Classes	Typical Characteristics	Disposal Options
1. Exempt waste (EW)	Activity levels at or below clearance levels given in Ref.[18], which are based on an annual dose to members of the public of less than 0.01 mSv.	No radiological restrictions
2. Low and intermediate level waste (LILW) 2.1 Short-lived waste (LILW-SL) 2.2 Long-lived waste (LILW-LL)	Activity levels above clearance levels given Ref. [18] and thermal power below about 2kW/m ³ Restricted long-lived radionuclide concentrations (limitation of long- lived alpha emitting radionuclides to 4000 Bq/g in individual waste packages and to an overall average of 400 Bq/g per waste package) Long-lived radionuclide concentrations exceeding limitations for short-lived waste	Near surface or geological disposal facility Geological disposal facility
3. High level waste	Thermal power above about 2kW/m ³ and long-lived radionuclide concentrations exceeding limitations for short-lived waste	Geological disposal facility

TABLE IX. Potential sources of environmental impacts and mitigation

Factor	Mitigation
<p>Mining & Milling</p> <p>Temporary land disruption SO_x emissions Silica dust Radon & Radon daughters Heavy metals NH₃ Acid production Tailings</p>	<p>reclamation/proper practices scrubbers ventilation/filters ventilation, water or soil covers effluent treatment removal neutralization/anoxic covers tailings management facility designed to retard migration of radionuclides and reduce radon emissions</p>
<p>Refining and Conversion</p> <p>Temporary land disruption Low activity solid waste NH₃ Acids HF + F₂ Organics TBP + kerosene CaF residue</p>	<p>reclamation disposal in waste management facility recovery proper practices collectors + scrubbers/neutralization proper practices disposal in waste management facility</p>
<p>Enrichment</p> <p>Temporary land disruption SO_x + NO_x from electricity generation Contaminated water (anions and some metals) Sludges</p>	<p>reclamation flue gas treatment dilute and discharge retention</p>
<p>UO₂ Fuel Fabrication</p> <p>Temporary land disruption HF Liquid effluents (lower negligible activity) NH₃ + nitrogen compounds Strong acids Decommissioning</p>	<p>reclamation scrubbers and filters neutralized proper practices conventional techniques</p>

TABLE IX (cont.)

Factor	Mitigation
<p>MOX</p> <p>Temporary land disruption Atmospheric discharge Liquid effluents (low or negligible activity 1 - 5 kBq/m³) Liquid wastes of low or intermediate activity Solid waste</p>	<p>reclamation filters monitored and discharged</p> <p>conditioned to solid waste</p> <p>incinerated or compacted then encapsulated in concrete for waste management</p>
<p>Spent Fuel Storage</p> <p>Temporary land disruption Waste effluents Atmospheric discharges</p>	<p>reclamation concrete moulds filters</p>
<p>Spent Fuel Disposal</p> <p>Not yet an option</p>	
<p>Spent Fuel Reprocessing</p> <p>High level liquid waste Intermediate and low level waste Liquid effluent</p> <p>Atmospheric discharges Conventional chemicals and hazards</p>	<p>concentration and vitrification concentrated and converted to solids monitored filtered ion exchange scrubbers and filters plant design and proper practices</p>

3. TECHNICAL DESCRIPTION AND REVIEW OF RELEASES AND IMPACT OF NUCLEAR FUEL CYCLE FACILITIES

3.1. GENERAL

The following section discusses in very general terms the releases from the facilities of the nuclear fuel cycle. In Appendix A there is an evaluation of the operational impacts of both mines and mills. In addition the reader is referred to the DECADES database for additional information on the facilities including release data.

Mining and milling of ores involves the disruption of land surface and water bodies therefore the environmental impacts are potentially more numerous and diverse. The discussion of this aspect of the nuclear fuel cycle has been given more emphasis than other portions of the fuel cycle because of its nature.

3.2. MINING

3.2.1. Basic process

Uranium is widely distributed in the earth's crust and oceans where it has an average abundance of two parts per million (ppm) and five parts per billion (ppb), respectively. The average concentration required for economic recovery depends, among other things, on the market price of uranium. In recent years the uranium market price has been severely depressed and only higher grade deposits capable of lower cost production have continued to operate. Historically, uranium has been economically recovered using conventional production where ores contain average grades of about 0.1% uranium or more. In most cases in the western world where average grades are between 0.01% and 0.1% uranium (i.e. 100 to 1 000 parts per million (ppm)), uranium is recovered as a by-product of other mineral commodities. In Canada, deposits with up to 20% uranium are being developed.

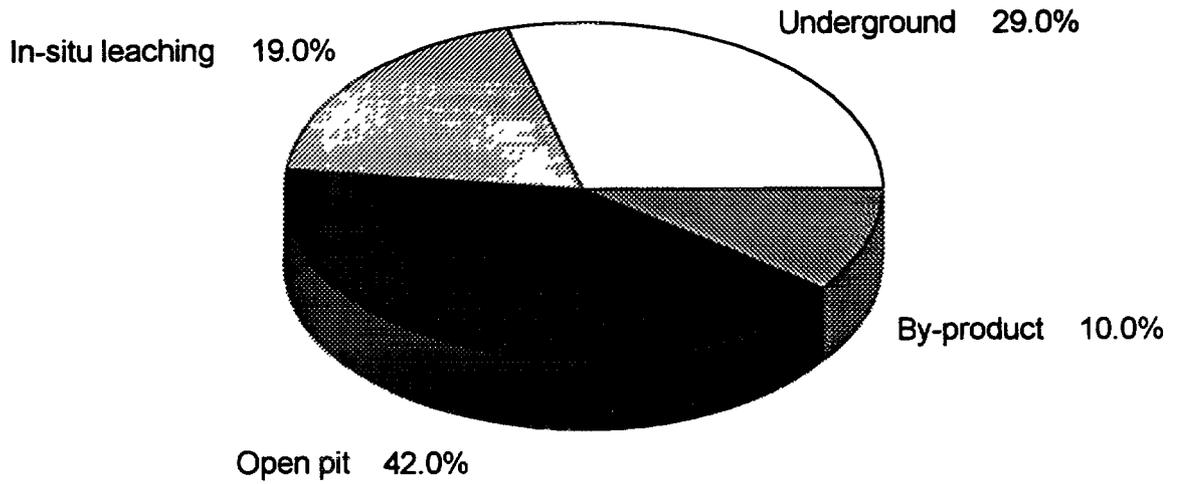
The ores containing uranium bearing minerals are usually mined by conventional open pit or underground methods depending on the geological condition of the ore, such as deposit size, ore grade, depth and ground condition. In general, the open pit method is employed when the ore body lies close to the surface under an overburden which can be removed easily and economically. Underground mining is typically used for ore bodies at depths greater than 100 m. This operation produces less waste rock than open pit mining. Conventional mining produced 71% of the world's uranium in 1993, 42% by open pit and 29% from underground mines [Uranium Institute (UI) data].

Some non-conventional methods, such as in situ leaching (ISL) and heap leaching, are also used for uranium production. ISL mining requires a porous ore body (sandstone) saturated with groundwater and confined between relatively impervious layers. The ore is left in the ground and a leaching solution (either alkaline or acidic, plus an oxidant) is injected into the ore body through wells. The solution percolates through the ore where it oxidizes and dissolves the uranium. The uranium bearing solution is then recovered by pumping. Uranium is extracted from the solution in a surface facility using ion exchange technology similar to those methods employed in some conventional ore processing plants. ISL mining produced 19% of 1993 world uranium production [UI data] (Fig. 6).

In most cases alkaline ISL leach systems, using bicarbonate solutions and dissolved oxygen as an oxidant, are considered to be more environmentally acceptable than are conventional mining operations. However, ISL mining can result in significant groundwater contamination, if a sulphuric acid system is used. Groundwater restoration following acid ISL mining is much more difficult to achieve.

Heap leaching is a similar process whereby broken ore is leached, either underground or on surface on an impervious membrane, by percolating sulphuric acid through the heap and recovering the uranium solution. Heap leaching is usually applied to low-grade ore produced from a conventional mining operation, which could not be economically milled. Currently, the amount of production from heap leaching is minimal.

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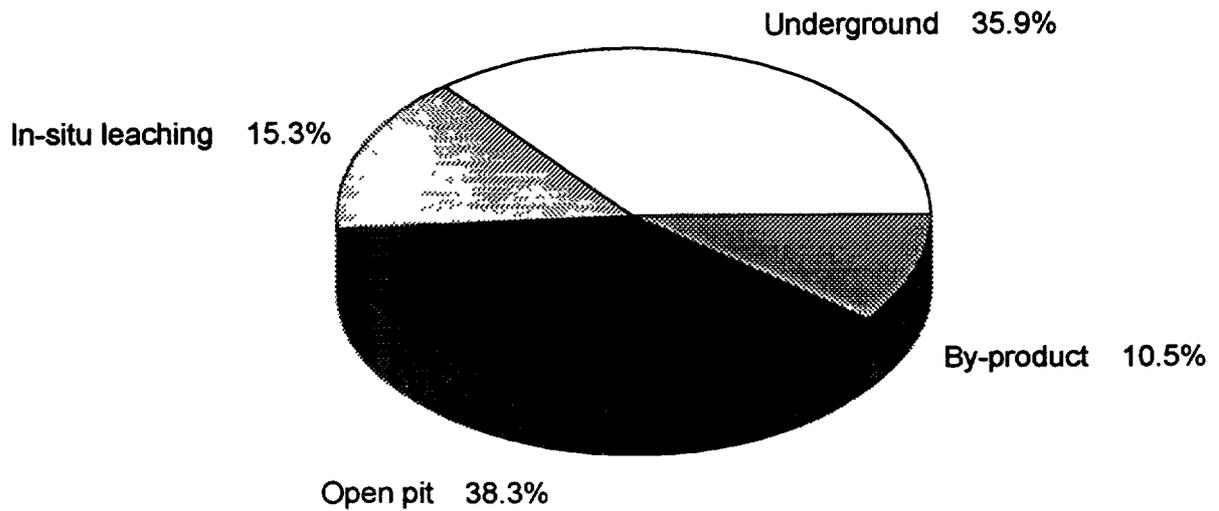


FIG. 6. World uranium production by mining methods

In addition, uranium is recovered as a by-product of other mineral processing, such as phosphate fertilizer as well as mining of gold and copper etc. This represented about 10% of 1993 world production [UI data].

3.2.2. Resource requirements

The quantity of ore required to produce the nuclear fuel depends on the average grade of the ore, which in the past has ranged between 0.1% and 2.5% U. Production of 1 GW(e) from a LWR requires about 200 tonnes uranium annually which corresponds to a mining rate of about 8 000 to 200 000 tonnes ore. The higher grade deposits now being developed would require a much lower mining rate.

Temporarily committed land use is currently estimated to be 25ha per 1 GW(e).a for open-pit mining [4]. However, this figure is expected to decrease as lower grade mines are exhausted and higher grade mines are developed. About one hectare of land is permanently committed [4]. During open-pit mining operations, earth overburden above the ore-body and barren rock are produced. The quantity of the waste rock removed is estimated to be around 10^6 tonnes per GW(e).a [2].

Underground mining requires the disturbance of relatively small areas of land, primarily for waste rock piles. During ISL mining, areas of land are used only temporarily and the surface disturbance from the operation is small.

Open pit mining produces a greater disturbance to the local area than does either underground mining or ISL mining. Disturbances to the land surface must normally be remediated following open pit mining and, consequently, the amount of land permanently committed is likely less than 1 ha per GW(e).a.

3.2.3. Releases

The releases from Uranium mining, for the most part, are similar to releases from conventional mines.

i) Radon and Radon Daughters

Radon and radioactive dusts are released to the atmosphere when an ore body is exposed and when it is broken during mining operations. Short lived radon progeny, resulting from the decay of radon, are a major source of radiation exposure for uranium mine workers particularly in underground mines. Ventilation is used in underground mines to remove radon and thereby limit the exposure to its progeny. However the exhausting of the radon and its progeny from underground mines results in a dispersal of these radionuclides into the environment.

At ISL mines radon gas is dissolved in the uranium bearing solution that is pumped from the ore body and may be released if the solution is exposed to the environment as in tanks or ponds. The average radon release in mining, normalized to the generation of electrical energy, is 75 TBq per GW(e).a [6].

ii) Contaminated Water

Water contaminated with radioactive or other materials is produced by the dewatering of underground and open pit mines, surface water runoff from and seepage through the waste rock piles and ore stockpiles, and water from ISL restoration activities. The radioactivity of this water is generally derived from the dissolution of soluble uranium, thorium, radium and lead ions. The water may also be contaminated with various heavy metals such as arsenic, selenium, and nickel. In cases where pyrites are present in the ore, the generation of acid requires neutralization of this water as part of the treatment process if it is discharged to the environment at this point. Acid generation is a concern of all types of mining in that the acid solubilizes and increases the mobility of the heavy metals, for uranium mining it also mobilizes the radionuclides. Blasting may also add some nitrites, nitrates, and ammonia to the mine water.

In many cases, contaminated water is collected and may be used in the mill process. This presents a means of reducing the amount of water contaminated, and recovering a small amount of uranium that may be present in this contaminated water. Another outcome of the recycling of the minewater is that a) only one treatment facility is needed, and b) only one point of discharge is required, thus simplifying the monitoring of the releases. In the treatment facility the effluent is neutralized where necessary, and chemicals and flocculants added. The treatment facility will comprise one or more evaporation pond through which the treated effluent will be passed to allow the precipitation of contaminants before release to the environment.

iii) Dust

Dust originating at exposed ore stockpiles, orehaul roads, may require the application of covers or, in the case of roads, watering to reduce the dust.

iv) Other

As in any other industrial undertaking, there will be possible releases of other substances. For example, fuel oils, contaminated solid wastes, regular landfill material. Where these are contaminated with radionuclides, they are disposed of to the tailings area, if not then appropriate landfill arrangements are made. Local environmental regulations and requirements will cover these situations.

3.2.4. Decommissioning

i) General

After closure of the operation, some form of decommissioning is required by most jurisdictions. This results in much of the land being returned to nearly its original condition. In addition, equipment and buildings must, typically, be removed and disposed of in an appropriate manner. Decommissioning requirements generally include restoration of aquifers involved in ISL mining.

ii) Mine

At the end of mining it is usual that some mineralization may be exposed in the walls and floor of an open pit, and this can give rise to ongoing release of contaminants such as radon, other radionuclides, and heavy metals. However, it is common for mined-out pits to be flooded or some other form of remedial action to be taken to limit these releases. In some cases, mined-out pits are used for mill tailings disposal. In most cases, release of radon from an underground mine is much smaller after mining ends since there is no longer forced ventilation, the mine floods and mine openings are sealed.

At ISL mines, the potential exists for contamination of adjacent aquifers. The requirements for natural physical barriers (i.e. geological confining layers) plus mandated monitoring makes adjacent aquifer contamination unlikely. Any such contamination that might occur can be relatively easily remediated.

If the open pit or underground working extends below the water table, groundwater must be removed to permit mining operations. This can result in temporarily lowering the water table. The effect is generally limited to the immediate vicinity of the mining operations. The water table returns to its normal level after pumping is discontinued.

iii) Waste rock piles and ore pads

As part of decommissioning, wastepiles which could be a source of heavy metal and radionuclide contamination are generally recontoured and vegetated to limit water infiltration, radon exhalation and wind erosion. In some cases some of the underlying material may be removed to the tailings area if a mill is part of the operation.

iv) *Treatment ponds*

The precipitant resulting from the treatment will have to be disposed of in an appropriate manner at decommissioning as they can form a possible source of future contamination. One possible way is to remove the precipitant to the tailings area.

3.2.5. **Accident considerations**

The most likely accident in uranium mining operations that may have environmental consequences is the release of contaminated water from a pipeline. In general, monitoring systems are in place to ensure that such releases are detected quickly, in order that the consequences may be limited and remedial action undertaken.

3.3. **MILLING**

3.3.1. **Basic process**

Uranium ore is generally processed close to the mine to limit transportation costs. The typical process for the extraction of uranium consists of crushing and grinding of the ore, followed by chemical leaching with sulphuric acid or an alkali-carbonate solution. Acid leaching is the more common method; however, some mills use alkaline leaching when the ore body contains limestone or similar basic constituents which would consume uneconomic quantities of acid. The uranium solution is purified and concentrated by ion exchange, and/or solvent extraction technology. The uranium is then precipitated from solution, filtered and dried to produce a concentrate, known as yellowcake, which contains between 60% and 90% uranium by weight.

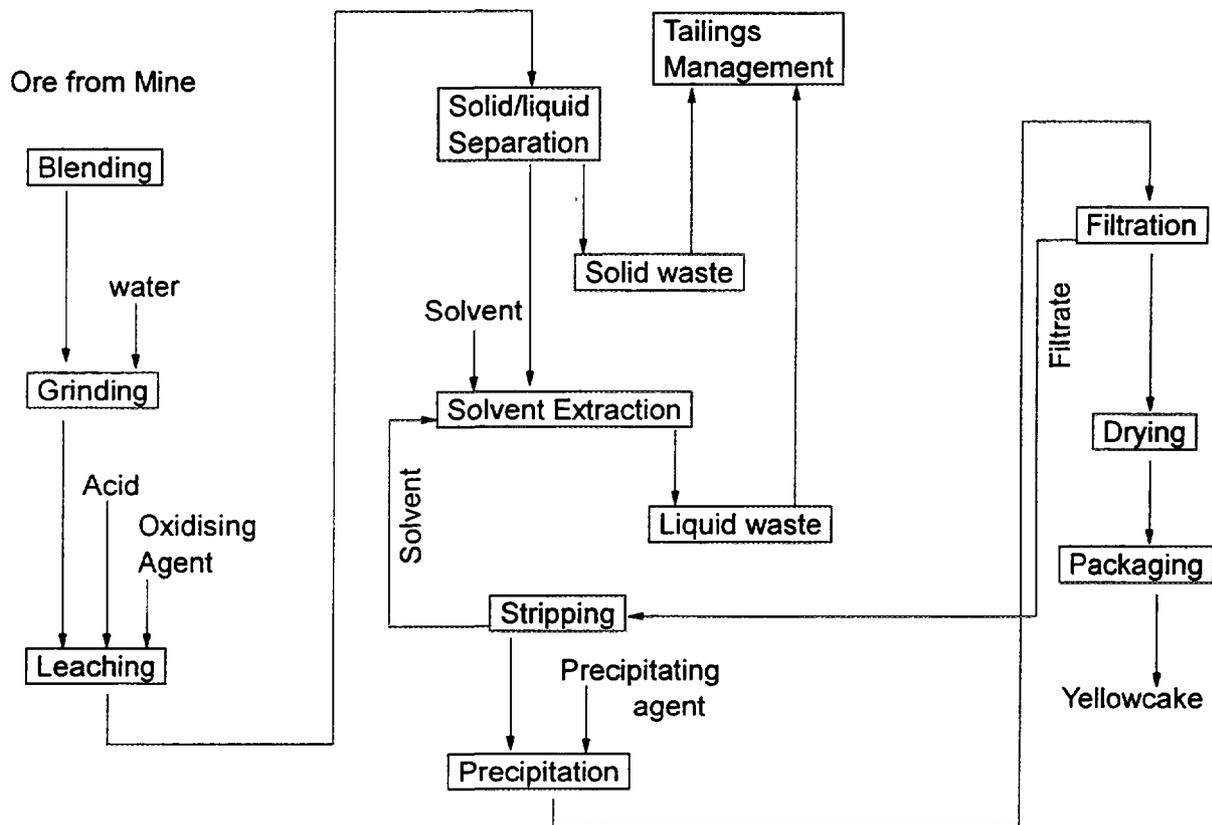


FIG. 7. *Typical simplified flow sheet for acid leach uranium mill*

- The basic steps in the acid-leach solvent extraction process, shown in Fig. 7 are:
- a. Ore is blended to give a consistent grade of mill feed, then crushed and ground to provide a large surface area for the subsequent chemical processing. It is mixed with water to form a slurry to facilitate the grinding and in order that it may be transported through the circuit.
 - b. The slurry is leached with sulphuric acid and an oxidizing agent which leaves the uranium in the liquid phase.
 - c. Liquid/solid separation then takes place. The solid phase is waste, known as tailings, which is transferred as a slurry to the tailings management facility.
 - d. The product liquor is pumped to a solvent-extraction circuit where uranium transfers from the aqueous phase to the solvent phase. (At some facilities, ion exchange is used instead of solvent extraction.) The aqueous phase (raffinate) is waste, which is usually blended with the tailings for disposal.
 - e. The uranium is stripped from the organic solvent and then precipitated as a chemical compound. The organic solvent is recycled.
 - f. The uranium bearing compound (yellowcake) is then dried, and may also be calcined, before being packaged for shipment.

In ISL processing the uranium bearing solution from the mining operation is fed directly to an ion exchange circuit. Some facilities then use solvent extraction to further purify the uranium concentrate. Precipitation, drying and packaging at an ISL facility are similar to those in a conventional mill.

3.3.2. Resource requirements

It is estimated that currently an average of about 4 ha of land area are required per GW(e).a for milling [4]. About 75% of this land is devoted to an impoundment for the permanent disposal of mill tailings. The amount of land required is highly dependant on the grade of ore being processed. As lower grade mining and milling operations cease, and are replaced by higher grade facilities, the area of land used per GW(e).a will decrease significantly.

3.3.3. Releases

i) Tailings

The tailing slurry is the most significant waste from the milling process. This stream is a mixture of leached solid ore and waste solutions from the grinding, leaching, uranium purification, precipitation and washing circuits of the mill. Because of the small initial content of uranium, the tailings are essentially of the same volume as the feed supply of ore. About 4×10^4 to 6×10^4 m³ of uranium mill tailings are produced per GW(e).a [10].

The tailings are characterized by their relatively large volumes and relatively low activity concentrations of long-lived natural radionuclides. About 15% of the total radioactivity which was originally contained in the ore is retained in the yellowcake produced by the mill. Once the shorter-lived radioactive nuclides have decayed, some 70% of the radioactivity originally present in the ore is left in the tailings. The tailings contain nearly all of the naturally occurring radioactive daughters from the decay of uranium, notably thorium-230 and radium-226. The presence of thorium-230 provides a long-term source of radon emission.

The tailings generally will also contain heavy metals, which were present in the original ore, but they will also have incorporated process chemicals such as ammonia and organics. The tailings represent a long term source of these substances into the environment through migration in the groundwater below

the tailings. The impacts of these long term releases must therefore be quantified and assessed. This is usually done by monitoring and predictive modelling.

By its nature, milling is designed to change the mineralogical and chemical characteristics of mined ore and put the desired uranium product into a more soluble, concentrated form. Such milling operations also tend to increase the solubility of certain contaminants associated with the ore. For example, the acid process tends to result in dissolution and potential mobilization of radium decay products, and various heavy metals present in the ore. Tailings management facilities are designed to control the release of these contaminants and/or mitigate their consequences.

ii) Contaminated water

Mill sites in dry areas give rise to effectively no liquid effluents. However the runoff water of mills in wet climates will contain radionuclides and may need treatment before release into watercourses.

Contaminated water is discharged from uranium mills to tailings management facilities. The contaminants will include radionuclides, heavy metals, sulphates, chlorides, organics and ammonia. The exact mixture will depend on factors such as process and ore grade. Treatment of the water will reduce the loadings of heavy metals, radionuclides and some anions. However monitoring of the released water to ensure compliance with regulatory limits on all contaminants will have to be done.

Most of the water either dissipates through evaporation or is treated and discharged to the environment (see discussion section 3.2.3). In some cases the water may be recycled to the mill further reducing the total amount of water contaminated.

iii) Airborne releases

Radioactive airborne effluents from milling may include dusts and radon gas released into the air from ore stockpiles, crushing and grinding of ore, drying and packing of yellowcake, and from the tailings retention system. The releases of dusts produced in the processing operations are reduced by ventilation extract scrubbers. Tailings may be a continuing source of radon and radioactive dust after milling operation has ceased. UNSCEAR [6] estimated the release rates of Radon-222 from mill, mill tailings during operation and abandoned mill tailings to be 3 TBq, 20 TBq and 1 TBq per GW(e).a, respectively.

Airborne chemical contaminants released to the environment include combustion products (oxides of carbon, nitrogen and sulphur) from the process steam boilers, power generation, sulphuric acid fumes in small concentrations from the leach tanks, and vaporized organic reagents from the solvent extraction ventilation system. In addition, some plants where sulphuric acid is made on site, sulphur dioxide is exhausted to the atmosphere.

3.3.4. Potential Health and Environmental Impacts during routine operation

i) Occupational

Radiological

The exposure to workers in the mill may result from the ore dusts in the crushing and grinding areas, from exposure to short-lived radon progeny and gamma radiation where ore and tailings are handled, and from the yellowcake dust in the precipitation, drying and packing areas. UNSCEAR [6] evaluated the annual average effective dose per monitored worker to be 6.3 mSv. Internal exposure is the greatest contribution to total exposures in milling. Its figures showed about 38% of exposures arose from the inhalation of radon progeny, about 47% from inhalation of ore dust and about 15% from external irradiation.

Non-radiological

Workers in uranium mills are exposed to hazards that are similar to those encountered in any chemical processing facility or mill, including industrial-type accidents. Reagents used in the extraction process and moving machinery are possible causes of worker injuries.

ii) *Environmental*¹

Radiological

UNSCEAR [6] evaluated releases from the milling stage, as shown in Table III. The normalized collective effective dose shown is based on a model with a population density of three people per km² in the vicinity of mills. However, much of the world's uranium is produced in areas with a population density lower than this and the doses are, therefore, likely overestimated.

Potential sources of hazards to public health and the environment from mill tailings comprise the following types of release: 1) escape of gaseous radon, 2) transport of radioactive particulates by the wind, 3) radionuclides, heavy metals, or other toxic materials may contaminate surface water and groundwater through runoff or seepage; and 4) tailings may be dispersed over a wider area by erosion or flooding.

Non-radiological

Impacts of mining activities are measured using various standards. For radiological impacts to humans the criterion is dose. However, for non-radiological and radiological impacts on the environment excluding humans, several other criteria are used. Frequently baseline studies are done before any mining activities are undertaken to establish environmental quality objectives to be used during the construction, operation, decommissioning and post decommissioning phases. In other cases there are pre-established air, sediment, water, and groundwater objectives that must be met. Social, aesthetic, economic and recreational values are met by establishing requirements to preserve parts of the affected ecosystem. For example, fish spawning areas and the habitats of endangered species may need to be preserved for economic or social and ecological reasons.

In many cases there is a requirement to meet toxicological criteria to ensure this level of environmental protection during the different phases of the undertaking.

The exact criteria will vary from site to site, and country to country. In the case of radiological impact, the pathways and diets will vary thus a generic statement of impact must be used with caution.

3.3.5. Decommissioning

After closure of the operation, some form of decommissioning is required by most jurisdictions. This results in much of the land occupied by the mill and associated buildings being returned to nearly its original condition. The equipment and buildings must, typically, be removed and disposed of in an appropriate manner.

After the mill and the associated tailings management facility have been decommissioned, control, surveillance, and maintenance of waste retention systems may be required for an extended period. Modern tailings management facilities have been designed with decommissioning in mind and address concerns with such things as the fluid nature of the slime portion of the tailings, difficulties of draining, covering

¹The topic of the environmental impacts of mill tailings has been exhaustively investigated as documented in such publications as: [11] "Scientific Basis for Risk Assessment and Management of Uranium Mill Tailings"

and revegetating the tailings mass, erosion of tailings and covers, seepages both into and out of the tailings management area, and the mobilization of heavy metals. The latter is a particular concern where acid may be generated in tailings resulting from the processing of ores containing pyrites.

Older facilities will have to address these concerns at the time of close-out.

Further information on uranium extraction technology may be obtained in IAEA Technical Report Series Nos. 359 [12] and 362 [1].

3.3.6. Accident considerations

The most likely types of accidents associated with uranium mill operations are inadvertent discharges of tailings to nearby rivers or streams or a major fire in a solvent extraction circuit. Inadvertent discharges would be caused by the failure of tailing dams. However, such failures may be minimized by appropriate siting, and preclosure stabilization.

Similarly should a contaminated water or tailings line break, the impact could be mitigated by appropriate design to channel or collect the spill flow.

The solvent extraction circuit, where solvent (mostly kerosene) and natural uranium are involved, represents a potential for a serious fire. Conventional safety measures are in place to prevent or reduce a fire accident.

3.4. REFINING AND CONVERSION

3.4.1. Basic process

Uranium Ore Concentrate (UOC) is an impure uranium compound, which may consist of any of several different uranium compounds. It still contains other elements than uranium and some of uranium's radioactive decay products. UOC is refined to produce pure uranium compounds. Two different processes - wet and dry - are used to purify the uranium and convert it to a usable form. The more common process, the wet process, consists of dissolving UOC in nitric acid and refining by solvent extraction. The pure uranyl nitrate is then converted to uranium trioxide, which can be converted to uranium dioxide for natural uranium fuel, or converted progressively to uranium tetrafluoride and uranium hexafluoride for enrichment to produce light water reactor fuel (Figs. 8a and 8b).

i) Wet process

The basic steps in the wet process are:

- UOC is dissolved in nitric acid;
- The resulting solution of uranyl nitrate hexahydrate is purified by solvent extraction. The aqueous uranyl nitrate solution is contacted counter-currently with an organic extractant, generally tributyl phosphate (TBP) diluted with kerosene or dodecane. Impurities are left in an aqueous raffinate;
- Concentration and thermal decomposition of uranyl nitrate to uranium trioxide (UO_3), with recovery of nitric acid; or concentration and precipitation as Ammonium diuranate (ADU) using ammonia. ADU is then converted to UO_3 by drying and calcination. The by-product is ammonium nitrate which is sold as a fertilizer;
- UO_3 is reduced to UO_2 with hydrogen;
- UO_2 is converted to uranium tetrafluoride UF_4 by reaction with hydrogen fluoride, HF;
- The solid UF_4 powder is transformed into the gaseous UF_6 with pure fluorine gas.

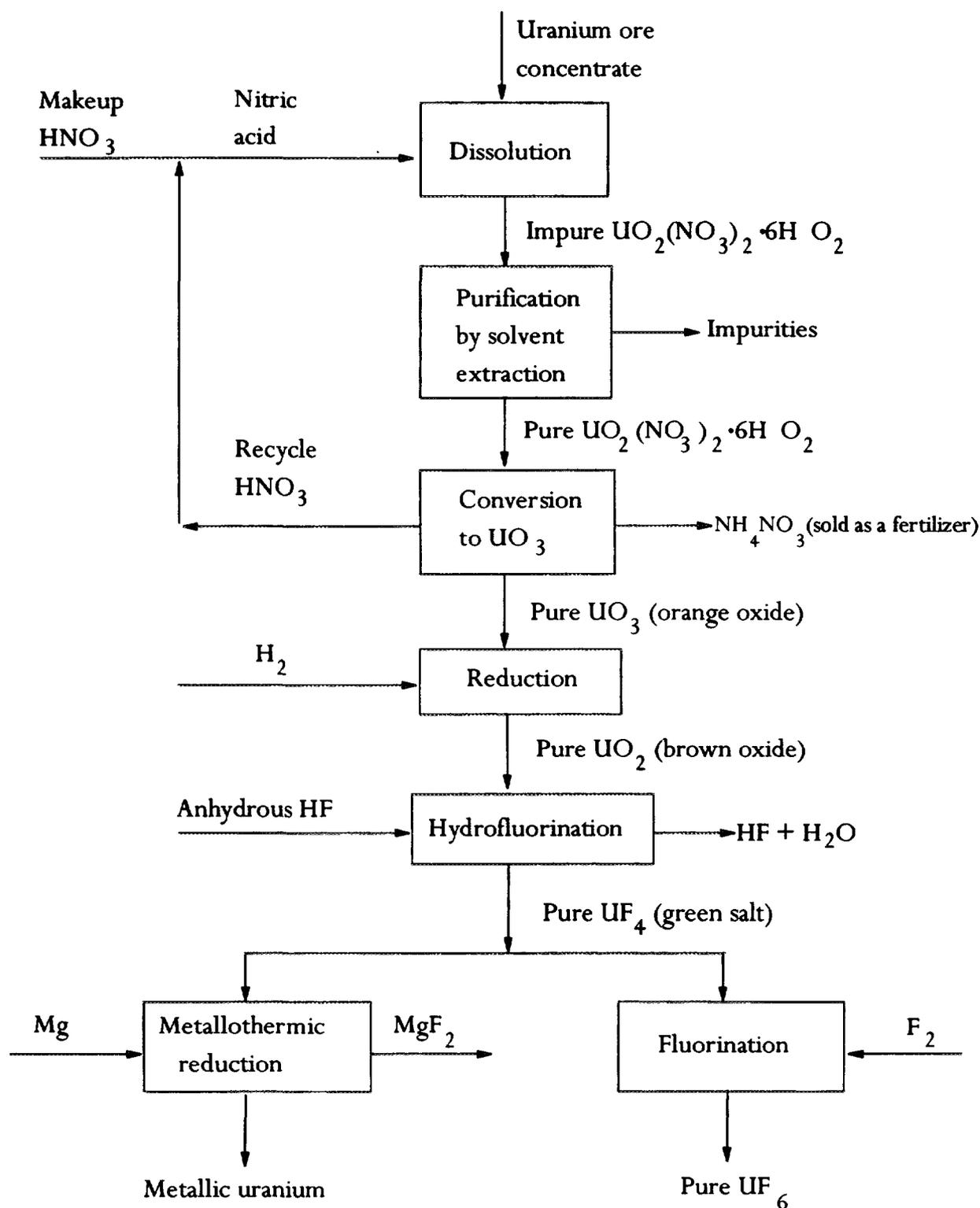


FIG. 8a. Flow sheet of conversion to UF₆ (Wet process)

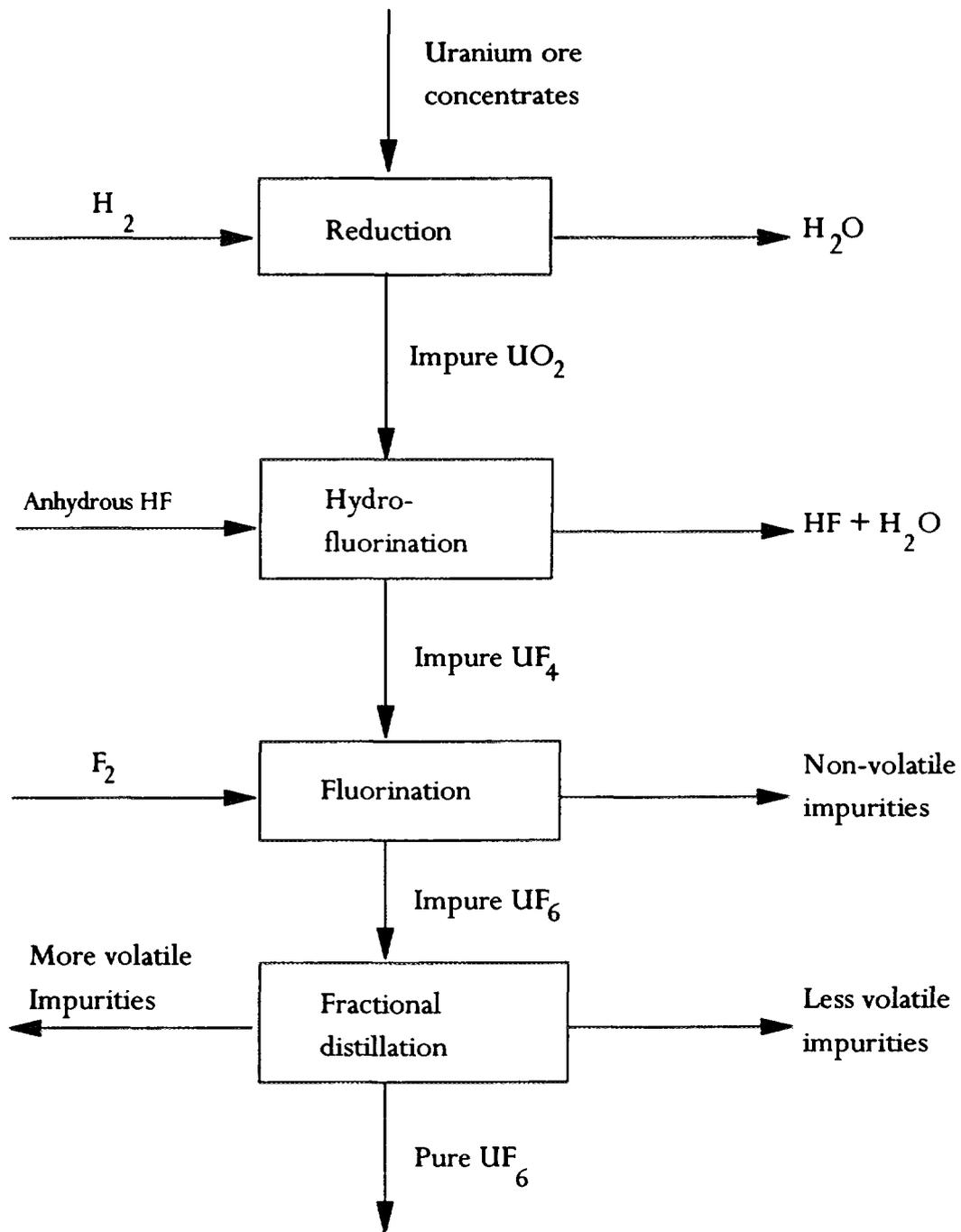


FIG. 8b. Flow sheet of conversion to UF_6 (Dry process)

ii) *Dry process*

The dry process converts UOC directly to impure hexafluoride, which is then purified by distillation. The main steps are:

- Treatment of UOC for processing in a fluidised bed (treatment processes may include crushing and pelletisation of UOC and, possibly, removal of excessive sodium impurity);
- U_3O_8 is reduced to UO_2 with hydrogen;
- UO_2 is converted to UF_4 by reaction with hydrogen fluoride;
- UF_4 is transformed to UF_6 with elemental fluorine;
- The crude UF_6 is fractionally distilled from other volatile and non volatile fluorides.

3.4.2. Resource requirements

Of the land commitment of hexafluoride production (about 1.3 ha per GW(e).a), approximately 10% is disturbed for roads, fills and plant structures. About 1% is permanently committed for waste burial [3].

3.4.3. Releases

The amount of water used with hexafluoride production is approximately 125,000 m³ per GW(e).a, about 90% of which is returned to the source from which it came (the water is used for process cooling) [3]. Some water is used as process water in the wet solvent extraction process. The remaining water is discharged to the air through evaporation from the holding ponds.

The effluent from the two methods of UF_6 production differs substantially. In the wet process most of the impurities entering with UOC are rejected in raffinate solution from solvent extraction, whereas in the dry process, most of the UOC impurities are contained in solid wastes from the fluorination and distillation stages.

Effluent from a typical wet process consists of:

- neutralized aqueous raffinate from solvent extraction. The types and amounts of impurities vary with the yellowcake source and represent only a few percent of the total product.
- caustic effluent and residual fumes from recovery of nitric acid, hydrogen fluoride and treatment of general off-gas streams, and
- a small quantity of solid calcium fluoride from the fluorination step.

The raffinate stream amounts to a few m³ per tonne of uranium processed. It may contain substantial dissolved solids, radium and thorium-230 entering with the UOC feed. It may also contain residual TBP and heavy metals. This stream is neutralized and impounded in a retention pond. It should be noted that the neutralization process may lead to elevated suspended solids loading in the effluents.

Disposal of this effluent from the raffinate stream, is a major problem associated with the wet process. Some facilities recycle raffinate to a uranium mill, where residual uranium is recovered and the remaining waste is disposed of with the tailings. Some scrubber effluents are treated with lime to precipitate fluoride ion in settling ponds for packaging and burial as calcium fluoride. In other cases the calcium fluoride is mixed with the flame reactor ash and recycled. Calcium fluoride is essentially insoluble thus its dissolution once buried is extremely slow.

Most of the chemical solid effluent from the dry process occurs as non-volatile ash containing iron, calcium, magnesium, copper and other fluorides. This residue can amount to about 0.1 tonne per tonne of UF_6 produced [3]. After recovery of remnant uranium, the final residue is packaged and buried as low

activity solid waste. Scrubber effluent arising from treatment of the hydrofluorination off-gas stream is treated similarly to those in the wet process.

3.4.4. Potential Health and Environmental Impacts during routine operation

i) Occupational

Radiological

The primary source of radiation exposure in uranium processing is the inhalation of uranium dust. This exposure pathway can be controlled by proper process design and operation to suppress and control uranium dust. Uranium hexafluoride is volatile and chemically reactive, but because of these properties it is always processed within a sealed system.

Through most of the processes, penetrating radiation is not a problem. However, in the conversion of UF_4 to UF_6 , uranium is separated from its short-lived progeny (^{234}Th and ^{234}Pa), leaving a small volume of high specific-activity material which emits penetrating radiation. However, with proper precautions, this material can be dealt with without significant personnel exposure.

An existing plant has normalized collective effective dose of 0.013 man Sv per GW(e).a.

Non-radiological

The refining and conversion of uranium entails the use of several strong chemical reagents. The most significant ones are nitric acid, sulphuric acid, ammonia, anhydrous hydrofluoric acid, and fluorine. Tributyl phosphate is generally used as a solvent, in a diluent such as kerosene. All of these reagents are in common use in the chemical industry and can be handled safely. Almost all uranium processing takes place within sealed systems, which eliminate personnel exposures except in upset conditions or maintenance work.

ii) Environmental

Radiological

Uranium dust

Emissions from uranium refining and conversion facilities are minor. The only radioactive one of significance is natural uranium, which may be emitted as dust or as a volatile reaction product of uranium hexafluoride. These plants are equipped with dust collectors and scrubbers to mitigate the impact of these emissions.

Raffinate

In the case where the material is recycled either through the mill circuits or disposed of to a uranium mill tailings facility, the radiological impact of the raffinate is included in the radiological impact of the ore facilities. In the case where the raffinate is not recycled the radiological impact is due to residual Th and to a lesser extent Radium.

Non-radiological

Atmospheric emissions from this process contain greenhouse gases such as NO_x . Aqueous releases may contain residual TBP and heavy metals that could have a potential impact on benthic fauna. Other contaminants with potential for environmental impact are nitrates and under accident conditions ammonia and fluorides may also be released.

3.4.5. Decommissioning

The basic technology for refining and conversion plants decommissioning is available proven and simple. The decommissioning technology is similar to that used for uranium mines and mills. Wherever possible, equipment and material is decontaminated and reused by other industries. If the material or equipment cannot be decontaminated it is disposed of to an appropriate waste management facility.

In the case of specialized equipment dealing with HF, practical experience has been acquired with the decommissioning of manufacturing equipment such as organic extraction facility, hydrofluorination reactor, fluorination reactor, cold trap etc.

Research and development continues for the decommissioning of ponds and to recycle the nitrates contained in the ponds as a fertilizer.

3.4.6. Accident Considerations

The largest potential accident in refining and conversion plants is chemical (e.g. accidental release of UF_6 due to a rupture of UF_6 cylinders and heavy damage to HF and NH_3 storage facilities). The radioactivity of UF_6 is very low, but when UF_6 is released to the air, it reacts with moisture in the air to form uranyl fluoride (UO_2F_2) and highly reactive hydrofluoric acid (HF). This reaction proceeds rapidly and liberates heat accompanied by a volume expansion. Preventive measures are taken to avoid such an occurrence or to limit the distance of the plume to such an extent that the public in the surrounding area is not injured.

3.5. ENRICHMENT

3.5.1. Basic process

LWRs require uranium enriched to 2-4% in uranium-235. Commercial enrichment technologies are based on gaseous diffusion or centrifugation of uranium isotopes in the form of UF_6 .

In the diffusion process, gaseous UF_6 is compressed and passed through a porous membrane. Molecules of UF_6 containing the lighter isotope, uranium-235, diffuse through the membrane more rapidly than those with the heavier uranium-238 isotope; consequently, the UF_6 passing the membrane has a slightly greater proportion of molecules containing uranium-235. The degree of enrichment for one membrane is minute and over a thousand successive diffusion stages are necessary to change the proportion of uranium-235 from the naturally occurring level of 0.71% in the feed stream to the approximately 3% required in the product stream for LWR fuel with about 0.25% in a reject 'tails' stream. Each stage requires recompression of the gaseous hexafluoride. Uranium enrichment by gaseous diffusion requires large quantities of electrical energy. About 40 MW(e).a of electricity would be needed by a gaseous diffusion plant to enrich the uranium for the generation of 1 GW(e).a of electricity in a LWR [3].

During centrifugation, molecules of UF_6 containing the heavier isotope uranium-238 migrate preferentially to the wall of a rapidly rotating cylinder. There is a consequent enrichment in the lighter uranium-235 isotope in the gas near the tube axis. The separation factor is greater than in the diffusion process, and two streams removed from the tube axis and wall require only tens of stages arranged in a cascade to produce the required percentages of uranium-235 in the product and reject tails. High centrifugal stress limits the size of the equipment, and many parallel cascades involving hundreds of thousands of centrifuges are required to achieve the separative capacity of a commercial enrichment plant.

3.5.2. Resource requirements

Essentially none of the land required for enrichment facilities is committed permanently. Temporary land requirement is estimated to be 0.3 ha per GW(e).a [2].

Water is required for the operation of cooling towers associated with the gaseous diffusion process and also for the large amounts of electricity which need to be generated. For 80% of the enrichment by gaseous diffusion, 92% of all the electricity required in the fuel cycle up to fuel fabrication was used in enrichment. Other enrichment techniques require less than one tenth of the electricity needed for a gaseous diffusion plant.

3.5.3. Releases

Emissions of radionuclides from the conversion, enrichment, and fuel fabrication processes are generally small and consist essentially of the long-lived uranium isotopes, ^{234}U , ^{235}U , and ^{238}U , along with ^{234}Th and $^{234\text{m}}\text{Pa}$, which are the short-lived decay products of ^{238}U . The long half-life of ^{230}Th prevents the activity build-up of any other radionuclide of the ^{238}U series.

The enrichment plant itself generates small quantities of airborne fluorides and oxides of nitrogen and sulphur from the process cooling systems, process cleanup operations, on-site steam plant and auxiliary production facilities. Uranium losses in the effluents are very low. Some sludges from container cleanup operations are retained on site. The depleted uranium residue from enrichment plants is normally stock-piled for possible future recovery of remaining fissile material. It is therefore not considered a release.

3.5.4. Potential Health and Environmental Impacts during routine operation

i) Occupational

Radiological

UNSCEAR [6] evaluated the radiological exposure to workers as follows. These doses are from external irradiation. Although the potential exists for internal exposure in enrichment plants, its contribution was negligible in comparison with external irradiation.

- Annual collective effective dose: 0.4 man Sv
- Annual effective dose per monitored worker: 0.08 mSv
- Normalized collective effective dose: 0.02 man Sv (GW(e).a)⁻¹

Non-radiological

Non-radiological effects on workers are minimized by following conventional safety procedures when handling equipment, chemicals, and other workplace hazards.

ii) Environmental

Radiological

Radiological impacts on the public are negligible. At the Capenhurst enrichment site in the United Kingdom, doses to the critical group members of the public are estimated to be less than 0.01 mSv per year.

Non-radiological

The concentration of gaseous and liquid effluent are below the range for which deleterious effects have been observed.

3.5.5. Decommissioning

Decommissioning of civil enrichment plants has not yet been done as all existing plants are still fully operational.

3.5.6. Accident considerations

The only uranium compound which is presently suitable for enrichment is UF_6 . The same preventative measures used in the refining and conversion process are applied to enrichment. Facilities are designed to leak-tightness and are equipped with a detection alarm system for UF_6 leakage.

The potential accidents associated with uranium enrichment activities are criticality and unintentional release of UF_6 to the atmosphere. Criticality incidents are unlikely to occur since uranium is handled for the most part in the gas phase at low enrichment, and due to the use of geometrically safe equipment and moderators.

3.6. UO_2 FUEL FABRICATION

3.6.1. Basic process

The feed material for the manufacture and fabrication of fuel for a LWR is uranium hexafluoride (UF_6) enriched to about 3% in uranium-235. The UF_6 is converted to uranium dioxide powder (UO_2) which is formed into pellets, sintered to achieve the desired density and ground to the required dimensions. Fuel pellets are loaded into tubes of Zircaloy (a zirconium-tin alloy) stainless steel, which are sealed at both ends. These fuel rods are spaced in fixed parallel arrays to form reactor fuel assemblies.

Several processes have been used for converting UF_6 to UO_2 .

- a. UF_6 is reduced to UF_4 with hydrogen, which is then hydrolysed by steam:
- b. UF_6 is converted first to ammonium diuranate (ADU) and then to UO_2 :
 - UF_6 , a solid at normal temperatures, is received in sealed cylinders from the enrichment plants
 - UF_6 is vapourised by heating electrically or with steam
 - The gaseous UF_6 is hydrolysed to form a solution of uranyl fluoride
 - Ammonia is added to precipitate ammonium diuranate (ADU)
 - The slurry of ADU is centrifuged or filtered
 - ADU is decomposed by heating, pyrohydrolysed with steam to remove traces of fluoride, and reduced to UO_2 powder with hydrogen.
- c. In the AUC (ammonium uranyl carbonate) process, streams of gaseous UF_6 , CO_2 , and NH_3 are fed into demineralized water, whereby AUC is precipitated. The AUC is converted to UO_2 by contacting it with steam and hydrogen with recovery of CO_2 and NH_3 (Fig. 9).
- d. UF_6 can also be converted to UO_2 by a dry conversion process. The gaseous UF_6 reacts in a vessel with H_2O steam to UO_2 and HF. The emission of HF to the environment is very small.

The fabrication operations involve the following stages:

- preparation of UO_2 powder of desired size distribution by comminution, compaction and granulation;
- manufacture of UO_2 pellets;
- sintering of the pellets in hydrogen gas;
- grinding of the sintered pellets to the required size;
- washing, drying and loading the fuel pellets into tubes of Zircaloy or stainless steel and sealing the ends with welded caps; and
- locating fuel rods in fixed parallel arrays forming the reactor fuel assemblies.

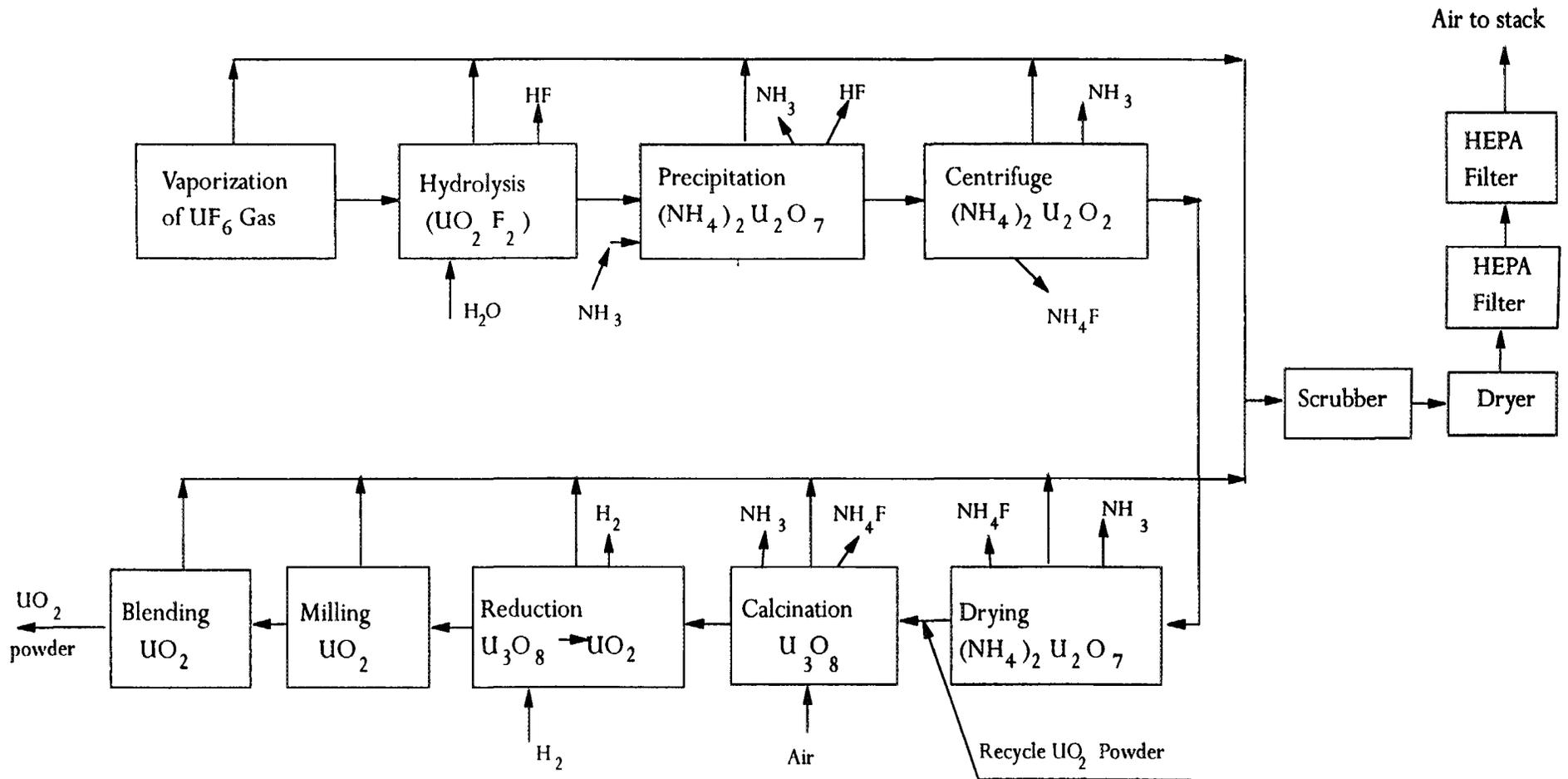


FIG. 9. Flow sheet of uranium dioxide conversion process (ADU process)

3.6.2. Resource requirements

Temporary land requirement is estimated to be 0.1 ha per GW(e).a [2]. All of the land required for fuel fabrication can be reclaimed by conventional techniques. Care would have to be taken in decommissioning the holding ponds or lagoons.

The facility requires water most of which is used for cooling of plant processes. It does not come into contact with uranium or process chemicals during operation. In the dry process, there is no cooling water discharge.

3.6.3. Releases

Radiological releases from fuel fabrication facilities were discussed earlier (see Section 3.5.3.) The effluent from fuel manufacture and fabrication with the greatest potential environmental impacts is chemical in nature. Hydrogen fluoride is potentially the most significant airborne chemical effluent from fuel fabrication.

Liquid effluent from fuel manufacture contains nitrogen compounds formed from ammonia in the production of UO_2 powder and by nitric acid in the scrap recovery operations. Very small quantities of uranium are released with the effluent gases and liquids. Ammonia and nitrates are found in liquids released from the waste holding ponds.

3.6.4. Potential Health and Environmental Impacts during routine operation

i) Occupational

Radiological

The exposure of workers in uranium fuel fabrication plants results from external exposure to gamma radiation emitted by the uranium isotopes of concern and their decay products and internal exposure from inhalation of uranium and its decay products. The external dose is very small, due to the low specific activity of uranium. The internal exposure depends on the design of the process equipment, especially the degree of encapsulation, i.e. the minimizing of airborne activity in the working area.

UNSCEAR [6] estimated the radiological exposure to workers as follows.

- Annual collective effective dose: 11 man Sv
- Annual effective dose per monitored worker: 0.5 mSv
- Normalized collective effective dose: 0.07 man Sv (GW(e).a)⁻¹

Non-radiological

Non-radiological effects on workers are minimized by following conventional safety procedures when handling equipment, chemicals, and other workplace hazards.

ii) Environmental

Radiological

UNSCEAR [6] estimated the normalized collective effective dose to be 0.003 man Sv (GW.a)⁻¹, with inhalation the most important pathway of exposure. The collective doses due to liquid discharges are much less than those from airborne discharges.

Non-radiological

In the dry process, the emissions of HF to the environment is very small. Nearly all of the produced HF will be removed from the off-gas by the condensing and cleaning system. The end products, liquid HF and $\text{CaCO}_3/\text{CaF}_2$, are suitable for industrial use. Thus the non-radiological impacts are mitigated by recovery and recycling of potential contaminants.

3.6.5. Decommissioning

Only conventional technology is necessary for the decommissioning of UO_2 production plants. Primary and secondary wastes generated are low activity.

3.6.6. Accident considerations

The accidents that could cause significant consequences are; a rupture of a heated UF_6 cylinder, an explosion in a reduction furnace and a criticality. Special design and operating precautions are taken to prevent the occurrence of those accidents.

UF_6 which is released into the vaporization room due to a rupture of the cylinder would be exhausted through a scrubber system and a high efficiency particulate air filter. The calcination of ADU to UO_2 powder is performed in furnaces with a reducing atmosphere of hydrogen and nitrogen. The hydrogen concentration is carefully controlled to prevent the development of an explosive atmosphere. The fabrication activity handles low enriched uranium under dry conditions. Hence the probability of the criticality accident is extremely unlikely.

3.7. MIXED OXIDE (MOX) FUEL FABRICATION

3.7.1. Basic process

Two processes are used to produce MOX fuel; these processes differ mainly in the beginning in the nature of the feed materials. For the dry process, feed materials are UO_2 (AUC, ADU, IDR) and PuO_2 powders. The alternative wet process starts with Pu- and U -nitrate solutions (Fig. 10).

Mixed oxide powder is prepared by co-milling or co-conversion, depending on the feed materials.

Further main fabrication steps (pelletizing, sintering, rod fabrication and assembling) are comparable with those of uranium fuel fabrication. The main differences from the UO_2 fuel fabrication are related to the strict alpha activity containment of the processed material in tight glove boxes and the shielding against gamma and neutron radiation.

A typical isotopic composition of plutonium originating from spent fuel is given in Table IV.

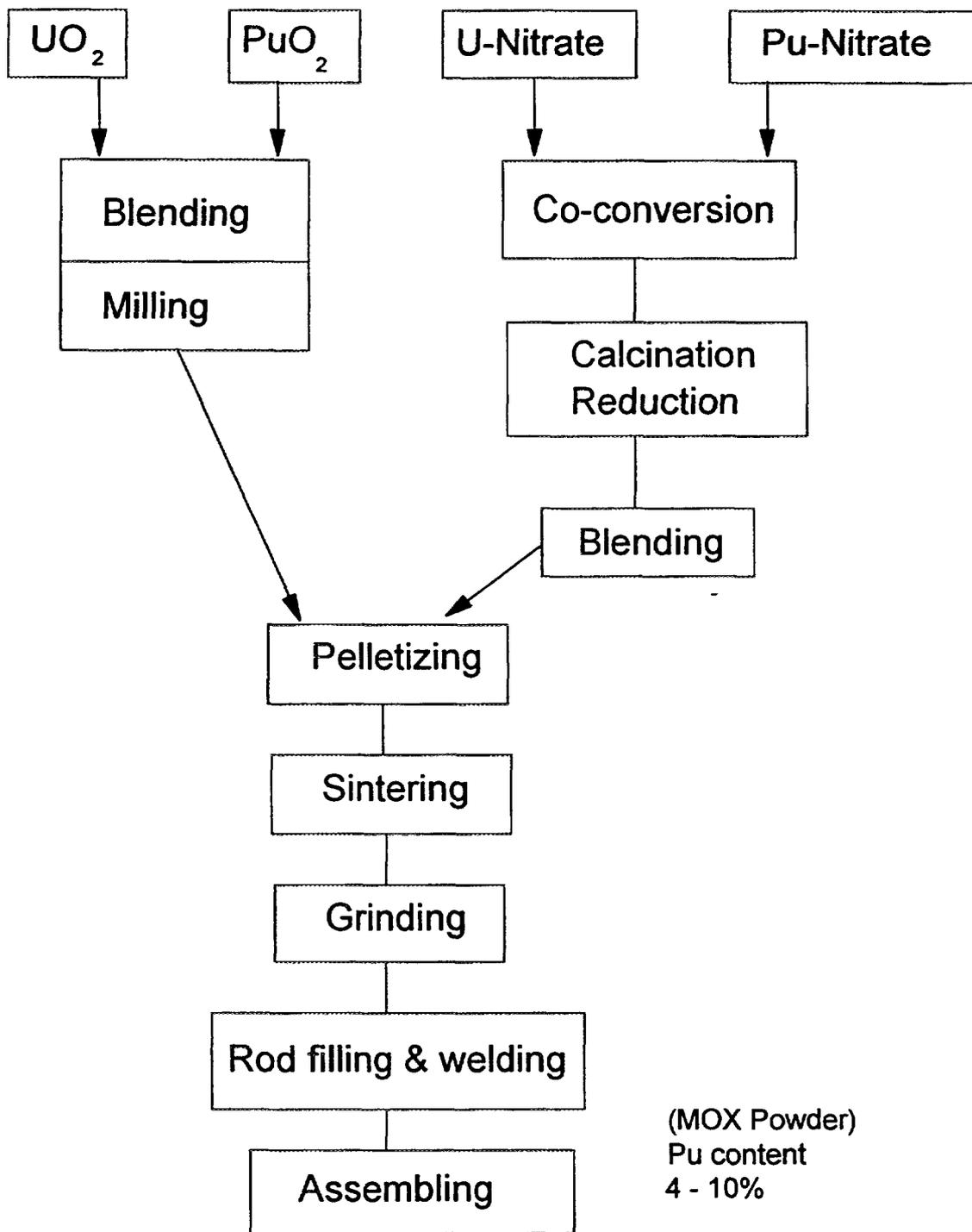
3.7.2. Resource requirements

Basically the MOX fabrication is a *recycling* activity.

The plutonium is a valuable raw material originating from the reprocessing of the UO_2 spent fuel. Replacing of U-235 by plutonium in the LWR fuel contributes to a better use of uranium resources.

In most cases for the matrix, natural uranium, depleted uranium, and reprocessed uranium can be used.

Uses of other resources (energy, fluids and land) are comparable with those in UO_2 fuel fabrication. Land requirement is estimated to be 3 ha per GW(e).a. The dry process does not use water and energy requirement is estimated to be 0.3 MW.a per GW(e).a.



(Dry process)

BELGONUCLEAIRE:MIMAS
BNFL:SBR
COGEMA:MIMAS.COCA
SIEMENS:OCOM

(Wet process)

PNC:MH
SIEMENS:AUPuC

FIG. 10. Flow sheet of MOX fuel fabrication

3.7.3. Releases

Effluents, both liquid and atmospheric, are produced by MOX plants. In addition, two streams of wastes are also produced.

Atmospheric discharge

Atmospheric discharge results from the ventilation - for dynamic containment purposes - of the production buildings and the glove boxes wherein the manufacturing process is performed. The discharge consists of aerosols of uranium/plutonium. Most of the discharged activity comes from the plutonium. Those discharges are mitigated by multiple absolute filtering.

Impact of atmospheric emissions is given in Becquerels for comparison purposes; (Table V) however conversion into effective dose to the public depends on specific site conditions such as dispersion conditions (wind intensities, landscape etc.) and dietary habits; the effective dose evaluation can also be influenced by national methodology (modelling etc.).

Resulting actual exposure to critical groups is less than 0.001 mSv/a, that is well below natural radioactivity.

The chemical emissions from the process are negligible (for the dry process, there is no chemical emission).

Liquid effluent

Very low alpha-active liquids principally coming from the cleaning of non or very low-contaminated areas (operators, floors etc.) are discharged in the environment after nuclear measurements (range of activity: 1 to 5 kBq/m³). Balance of the discharged activity is exactly maintained.

Liquid discharges are increasing with the throughput of the plants (these discharges are mainly depending of the number of workers and the surface of the buildings); typical discharged activity ranges from 0.5 MBq/a to 3 MBq/a.

Liquid wastes

Small volumes of low/medium activity solutions are generated in the process and treated as nuclear waste (for intermediate/final storage purposes, this liquid waste is conditioned into solid waste).

Solid waste

Solid waste generated in MOX fuel plants is either so-called suspect solid waste or slightly contaminated waste:

Suspect solid waste is composed of material which has entered the controlled area and is potentially contaminated; according to the characteristics of these wastes (burnable, compactible etc.), waste volume reduction can be performed before encapsulation for intermediate/final storage.

Contaminated waste is composed of material which has been in contact with plutonium; according to the characteristics (burnable, compactible etc.) and the level of contamination of these wastes, reduction of activity by decontamination and reduction of volume by incineration or compression can be performed; the waste is then conditioned for intermediate/final storage (for instance, by encapsulation in concrete).

The strategy of waste management is different from country to country, basically concerning the final storage.

3.7.4. Potential Health and Environmental Impacts during routine operation

i) Occupational

Radiological

The radiological impact to the operating staff results mainly from external exposure by gamma and neutron emitters. Internal exposure by incorporation of radioactivity is negligible during normal routine operation.

Indicative exposure figures are given in Table VI for existing plants under operation and for modern plants under construction; radiation protection management and compliance with the forthcoming new radiation protection regulations (ICRP 60 Recommendations) are based in the modern plants on enhanced mechanization and automatization.

Non-radiological

Non-radiological effects on workers are kept negligible by conventional safety measures when handling equipment, chemicals, or workplace hazards.

ii) Environmental

Radiological

The atmospheric discharge of radioactivity is extremely low due to the very highly efficient filtering of the discharged air.

Non-radiological

Non-radiological impacts to the environment are negligible (for the dry process, there is no emission).

3.7.5. Decommissioning

The basic technology for MOX plant decommissioning is available and proven (cleaning, decontamination, dismantling etc.). Nevertheless, research and development is still going on in some fields: new decontamination procedures and dismantling practices to reduce and minimize the radiation exposure and the generation of primary and secondary wastes.

Practical experience in the field has been acquired with the full decommissioning of plutonium laboratories and pilot plants and the decommissioning of manufacturing equipment in existing plants.

3.7.6 Accident considerations

Depending on the specific site conditions, the MOX fuel plants are designed and constructed to face most probable external hazards such as earthquake, flood, external fire, aircraft crash.

3.8. SPENT FUEL STORAGE

The following description is of a particular spent fuel storage system. There are other variations of this process. Furthermore the spent fuel storage facility is a closed system and generates atmospheric releases and ion exchange wastes that are then disposed of in waste management systems.

3.8.1. Basic process

A typical 1 Gw LWR discharges about 30 tonnes of spent fuel annually. When the spent fuel is removed from the reactor, it is highly radioactive and generates a considerable amount of decay heat. It is stored for one to three years in a water pool at the reactor site. Water serves as shielding and cooling medium to dissipate the heat released by fuel elements. Since "At the Reactor" spent fuel storage (AR) has limited capacity, fuel may be sent to "Away From Reactor" spent fuel storage (AFR) or reprocessing plant.

An AFR storage can be a facility by itself, or it can be part of another facility such as a reprocessing plant, a fuel cycle centre, or a disposal site where the spent fuel would be stored before emplacement in a permanent waste repository. In water-cooled AFR storage facilities, the spent fuel is stored in reinforced concrete pools filled with water. The surface of the concrete is covered with a water-impervious liner (stainless steel, epoxy or a combination of the two materials). Since the fuel arriving for storage in the AFR will have been cooled for at least a year at the reactor, the requirements in terms of cooling and cleaning systems are lower for AFR than for AR storage.

In addition to water cooled storage, dry storage facilities have been developed and constructed. In a dry storage facility, spent fuel is stored in casks or vaults, in a gas environment, such as an inert gas or air. A cask is a massive container which may or may not be designed to be easily transportable. When the transport cask is used, it minimizes the handling of the fuel itself. Vaults consist of above- or below-ground reinforced concrete buildings containing arrays of storage cavities suitable for containment of one or more fuel units. Dry storage has many advantages, e.g. the possibility of passive cooling, minimal or no maintenance, and a non-corrosive environment.

Whatever concept is chosen, there must be surveillance programmes in place to monitor the condition of the spent fuel. These programmes ensure that the fuel integrity is maintained. Such programmes have been in place for fuel that has been stored in water for several decades. The results of destructive and non-destructive analysis of the fuel pins show that no additional corrosion or degradation of the cladding has yet occurred.

3.8.2. Resource requirements

Large water-cooled AFR facilities have been constructed at La Hague in France (total capacity is 14 000 MTU - metric tonnes of uranium) and near the Oskarshamn nuclear power plant in Sweden (5 000 MTU) [9, 10]. CLAB, the central interim storage facility constructed in Sweden, comprises one above ground and one underground section. The storage section is located underground in a rock cavern. The cavern is 120 metres long, 21 metres wide and 27 metres high. It contains four storage pools and one smaller central pool connected to a transport channel. Each storage pool contains about 3 000 m³ of MTU. The CLAB facility was originally designed to store 3 000 MTU. By adopting high density storage racks, the storage capacity was increased to 5 000 MTU.

The spent fuel capacity of the THORP facility in the United Kingdom is 3 380 tonnes of LWR fuel, and 415 tonnes of AGR fuel.

3.8.3. Releases

Transport casks become contaminated both internally and externally in the course of normal operations and routine decontamination is necessary. The radioactive isotopes responsible for internal contamination are principally small particles of deposit becoming detached from fuel element surfaces (crud) and may be either fission products or transuranic elements from fuel-cladding failures. In addition, some contamination may arise from isotopes produced by activation of the non-fissile components of the fuel assembly; an important example of this is Co-60 produced by the activation of steel components.

In some cases, casks are loaded in ponds. If pond water is contaminated, this results in contamination of the external painted surfaces, mainly by the absorption of radiocaesium.

Decontamination is necessary for reducing surface contamination levels to comply with transport regulations and lowering operator dose during maintenance. Decontamination is carried out by pressurized water or chemical reagents. Pressurized water is recommended. Chemicals could lead to the generation of secondary wastes; in some cases this could result in mixed radioactive and hazardous waste. Chemicals may also have an adverse effect on the surface of a cask or its coating and make subsequent decontamination more difficult.

Nuclear fuel is surrounded by cladding which normally contains all the radioactivity in the fuel rod or pins. A very small number of cladding integrity failures does occur either in the reactor or subsequently. Failed fuel assemblies are stored in specially designed "bottles" that prevent contamination of pool water, especially by Cs-137.

Chemical control of the water in the pool is of paramount importance to ensure long-term integrity of the fuel, storage rack, water pool and auxiliary systems. The main parameters controlled include pH, Cl and conductivity. The pool water is also purified and cooled with heat and ion exchange units. Activity concentrations in pool water are kept at low levels, on the order of 7 (CLAB) - 18.5 (La Hague) MBq/m³. The experience at CLAB shows that the activity release to the storage pool water is to more than 95% ionic, 90-95% is Co-60 and the remainder is mainly Mn-54, less than 1% is Caesium.

The wastes that require management are the water and air effluents generated in the storage facility. They are treated in the same way as at the reactor. Concrete moulds are normally used to solidify spent ion exchanger resins, sludges and filters.

Further details of the releases may be obtained from the document [13], and in annex III "Environmental Safety of Reprocessing Plant La Hague".

3.8.4. Potential health and environmental impacts during routine operation

i) Occupational

Radiological

The collective radiation dose to CLAB staff and contractors was between 65 and 135 mman Sv for the years 1986-1993 [9]. At La Hague, the operator dose rate is less than 1 mSv/year/operator [14].

Non-radiological

Non-radiological effects on workers are kept negligible by conventional safety measures when handling equipment, chemicals, or workplace hazards.

ii) Environmental

Radiological

The radioactivity released to the environment has been kept low, hence impact to the environment is negligible. For example, at the THORP facility in the United Kingdom, the annual average dose to the critical group members of the public is in the order of 0.03 mSv.

Non-radiological

Non-radiological impacts to the environment are negligible.

3.8.5. Decommissioning

The same technology for decommissioning of spent fuel storage pool at reactor sites would be applicable to spent fuel storage facilities (cleaning, decontamination, dismantling, etc.).

3.8.6. Accident considerations

Accidental cladding failure, criticality, cooling system failure and loss of coolant are taken into account to design spent fuel storage facilities. Fuel-handling devices are designed to avoid accidental dropping or collision of fuel assemblies during transfer operations.

Criticality safety is guaranteed by the geometric configuration and the materials of the fuel storage rack having integral neutron poisons such as boron carbide. Criticality can result from deformation of the compartments in the fuel rack. Methods of preventing a criticality accident are the same as those used to prevent drops and shocks.

Cooling system failure is caused by power outage. To cope with this failure, backup diesel generators are equipped. Loss of coolant results from rupture of pool by an earthquake, or evaporation due to cooling system failure. Rupture of pool is avoided by adopting a seismic design. It will take ample time for water to evaporate. This time will enable the failure to be repaired or for arrangements to be made for external water supply e.g. by tank trucks.

3.9. SPENT FUEL DISPOSAL

3.9.1 Basic process

In the once-through option (Fig. 2), the spent fuel would be conditioned for final disposal after about 50 years of cooling. The fuel assemblies may be encapsulated directly or they may be disassembled using remote handling techniques so that the fuel pins can be packed together more closely. The encapsulated fuel in appropriate containers may be disposed of in deep, stable geological formations for an indefinite period of time. They may be embedded in a buffer material which will prevent any flowing ground water coming into contact with the container. Several geological media are under investigation.

The natural system of barriers comprises the repository host rock, the surrounding geological formation and the hydrogeological environment. There are two broad objectives: to minimize the probability that circulating groundwater will come into contact with the container and to minimize the migration of any radionuclides that may be released.

3.9.2. Resource requirements

Many factors must be considered in selecting a geological site for disposal. The site must be located in a region with a very low frequency of tectonic activity such as earthquakes and volcanism. The area should have a low potential for future exploitation for oil, natural gas or minerals. Despite these and other restrictions, it is likely that suitable sites can be found in most countries because many formations have been physically and chemically stable for millions of years and land requirements for disposal of spent fuel are not large. Refer to reference [5] for further information.

At this time, there does not yet exist any experience, on a commercial scale, of spent fuel disposal. Hence it is not possible to accurately predict the exact resource requirements for the construction, operation, and decommissioning of such a facility. However it is possible to state that the construction phase will have requirements and impacts similar to mining. Other requirements will be site and concept specific.

3.9.3. Releases

Because of the conceptual nature of spent fuel disposal it is not possible to quantitatively describe the impacts in this document. Currently the concept is under assessment in many countries. These assessments give consideration to the multiple barriers imposed between the fuel and the environment in modelling the impacts of the spent fuel disposal over periods of tens of thousands of years.

3.10 SPENT FUEL REPROCESSING

3.10.1. Basic process of Light Water Reactor

The spent fuel discharged from light water reactors contains about 0.8 to 0.9%, according to burn up, of uranium-235, 95% uranium-238, 1% plutonium and 3% fission products and other actinides. The uranium-235 concentration has fallen from around 3% to below 1% but is still on average above 0.7% that is present in natural uranium. The principal objectives of reprocessing are (1) to recover uranium and plutonium for reuse as nuclear fuels; (2) to remove radioactive and neutron-absorbing fission products from them; and (3) to convert the radioactive wastes in spent fuel into forms suitable for safe, long-term storage and disposal.

The difference between the once-through cycle and the reprocessing cycle is that the reprocessing cycle makes more efficient use of the fuel through extraction of Pu and recycling of the U235. This recycling can decrease the demand for natural uranium by up to 35%.

The spent fuel is first stored at the reactor pond in order to let its thermal power and its radioactivity decrease to a level acceptable for transportation to the reprocessing plant. Transportation is done in specially designed containers. On arrival at the reprocessing plant, the spent fuel is unloaded and stored again to allow its thermal power and its radioactivity to decrease to the design levels of the plant. Generally, for LWR fuels, total cooling time is at least three years (including 6 months at the reactor pond). The pond water becomes slightly contaminated primarily with radiocaesium, radiostrontium isotopes, cobalt-60 and manganese-54. Decontamination of this water is effected by filtration, to remove particulate, and by ion exchange to absorb activity present in soluble form.

The fuel is mechanically chopped into small pieces which are dipped into the dissolver solution. The fuel is leached from the chopped cladding by boiling nitric acid. After leaching, the fuel hulls (chopped cladding) are washed and transferred to storage.

The liquid in the dissolver will contain some insoluble material including fission product compounds from the fuel and both coarse and fine particulate from the cladding material, together with debris from the structural components of the fuel elements. These insoluble residues are separated by centrifugation or filtration. Both these wastes will need to be stored prior to disposal. The clear solution is analysed to determine its nuclear fuel content and is eventually adjusted for U-Pu concentration and acidity prior to extraction.

The liquor from the dissolver, after the removal of solids, is passed to successive extraction systems to separate the fission product wastes from the uranium and plutonium. The products from the solvent extraction cycles are uranium and plutonium nitrates, while the bulk of the fission products are removed as high level wastes for storage prior to conditioning and ultimate disposal. The solvent of the washing process is reused.

The two products of the solvent extraction process are first concentrated by evaporation. The uranyl nitrate solution can then be converted to uranium oxide form. Subsequently this product can be sent to another site for conversion to uranium hexafluoride and used in a re-enrichment cycle. The plutonium nitrate is converted first to plutonium oxalate by a precipitation process and subsequently to the oxide by thermal decomposition and calcination.

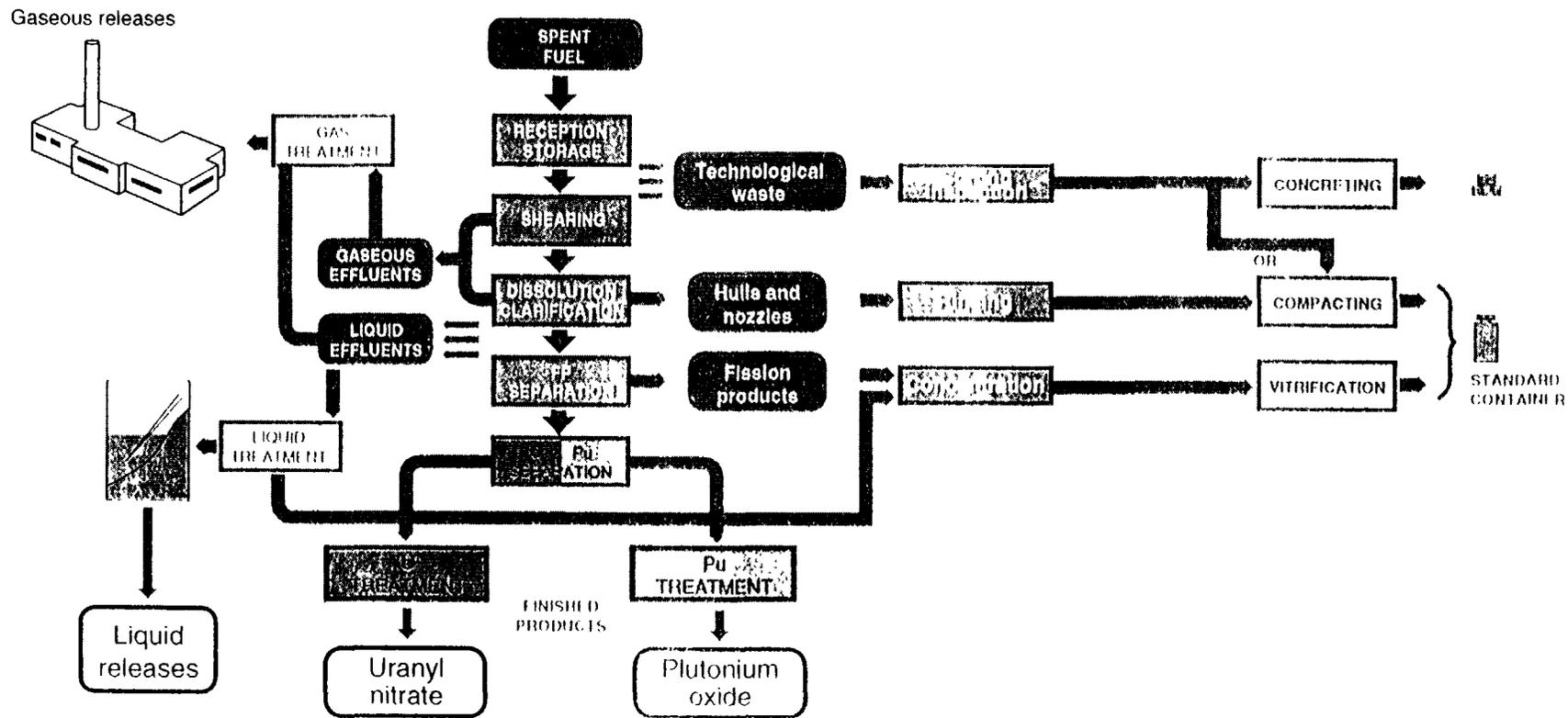


FIG. 11. Simplified flow sheet of the UP3 reprocessing plant

This process gives rise to several waste streams (Fig. 11); however since these are sent to waste management facilities they are not treated as effluent.

Radiological waste from reprocessing operations are classified into five categories:

- i) High level liquid waste
- ii) Intermediate level liquid waste
- iii) Low level liquid waste
- iv) Gaseous waste
- v) Solid waste

The first three liquid wastes are operationally classified on the basis of their activity levels. The distinction between intermediate and low level wastes is generally made on the basis of shielding requirements and on the extent to which they must be processed. Further details may be obtained in reference [15].

i) High level liquid waste

The high level liquid wastes are produced from the first cycle of fuel reprocessing. These contain the fission products and transplutonium elements, but also a very small residual fraction of uranium and plutonium. The wastes are concentrated by evaporation prior to storage in stainless steel tanks fitted with continuous cooling and agitating devices and afterwards vitrified. The glass is poured into stainless steel canisters which are stored for an interim period in air cooled vaults prior to final disposal.

ii) Intermediate- and low-level liquid waste

The intermediate-level wastes are all plant liquid wastes, including the aqueous raffinate from all stages of extraction in the plant other than the first cycle extraction, scrubber liquors, wastes from the highly active liquor evaporators and decontamination and plant wash liquors. These wastes are concentrated by evaporation and then held in tank storage till such time that they are converted to solid form for storage and disposal.

The low-level liquid wastes arise, after treatment, from the fuel storage ponds and from the chemical plant, where, for example, distillates from intermediate level wastes evaporators or supernates from the precipitation treatments of intermediate level liquid wastes, give rise to such effluents. These wastes are processed or recycled, so that most of the residues are in the high-level waste and the balance is either conditioned for disposal or is released as effluent to the environment.

iii) Gaseous waste

Some of the gaseous radioactive isotopes such as iodine-131 and xenon have short half-lives and decay in the fuel during storage. Other radionuclides such as tritium, carbon-14, krypton-85, iodine-129 have longer half-lives.

The feeds to the off-gas treatment systems consist of the dissolver off-gas stream and the general vessel off-gas stream that are routed through chemical and physical retention systems in the off-gas treatment trains.

The off-gases from shearing and dissolution of the spent fuel contain oxides of nitrogen (from the nitric acid) together with almost all of the krypton and xenon, a large fraction of the radioiodines, and a small part of the tritium formed in the fuel. Small amounts of semi-volatile ruthenium may also be present. In addition, particulates may be carried by the off-gases from the dissolver.

The gases are first passed through a condenser and a scrubber to remove nitrogen oxides and recover nitric acid which is recycled in the process. The residual gases then pass through several additional steps for removal of traces of NO_x, gaseous iodine (caustic scrubbers, solid sorbents) and

eventually carbon-14 and krypton-85. Finally, they pass through high efficiency filters before discharge to the stack.

iv) Solid wastes

If the solid wastes are not contaminated by plutonium or other transuranium (TRU) elements, they may be sent to an approved site for burial. Solid waste containing TRU elements include fuel-cladding hulls, particulate filters, discarded equipment and tools, and contaminated trash. Fuel cladding hulls can be embedded into a cement matrix inside containers for geological disposal. They can also be compacted and put inside stainless steel containers for geological disposal. Technological wastes, like discarded equipment or tools, are decontaminated in order to obtain a residual contamination level allowing a subsurface disposal. If the decontamination level remains too high, they can be compacted and embedded into a cement matrix for geological disposal. Burnable wastes contaminated with alpha emitters can be incinerated. Incineration ashes are decontaminated for plutonium recovery before being embedded into a solid matrix for deep disposal.

3.10.2. Resource requirements

Temporary land requirement is estimated to be 1.5 ha per GW(e).a [2]. About 95% of this land use is a fenced-in buffer zone and is not disturbed. La Hague reprocessing site is a 300 ha complex and its reprocessing capacity is 1 600 metric tonnes per year. It has 14 400 metric tonnes of spent fuel storage pools as well.

3.10.3. Releases

Radiological

The radionuclides of concern in reprocessing plant effluents are the medium and the long-lived nuclides, tritium, carbon-14, krypton-85, iodine-129, ruthenium-106, caesium-137 and isotopes of the transuranic elements. The important radionuclides which are released to the environment are tritium, carbon-14, krypton-85 and iodine-129. Data on normalized releases for both airborne and liquid releases from fuel reprocessing plants may be obtained from UNSCEAR [6] for earlier operations and from the European Union (EU) "ExternE project" for updated operations.

Non-radiological

All concentrations for elements released are monitored and are well below authorized limits, where satisfactory conditions of dilution exist.

3.10.4. Potential Health and Environmental Impacts during routine operation

i) Occupational

Radiological

UNSCEAR [6] evaluated separately the annual collective effective dose for the reprocessing shown in Table VII. The annual collective effective dose for reprocessing oxide fuel has increased from 0.36 man Sv to 5.7 man Sv reflecting the increasing amount of reprocessed fuel.

The average annual effective dose per monitored worker has declined from 4.0 mSv (for the period 1975–1979) to 1.4 mSv (for the period 1985–1989). Since publication of the UNSCEAR [6] data the new generation of reprocessing plants have achieved even lower levels of worker doses. For example, at Cogema's La Hague plant the average annual worker doses are now well below 1.0 mSv (0.26 mSv/man year in 1994).

Non-radiological

All reprocessing processes are operated remotely and take place within sealed systems, which eliminate personnel exposures except in upset conditions or maintenance work. Non-radiological impacts to workers are negligible.

ii) *Environmental*

Radiological

UNSCEAR 93 [6] gives the total average normalized collective effective dose as 0.05 man Sv per GW.a from airborne effluents and 0.2 man Sv per GW.a from liquid effluents in the local and regional domain. The annual effective dose to the member in the critical group of the public varies between .01 - .05 mSv. The EU "ExternE project" gives some data for updated operations.

The most significant contribution to the global category is the dose from C-14. A French study [10] shows that the average individual dose due to the natural occurring atmospheric C-14 is .012 mSv and the dose from C-14 due to the total annual production of nuclear energy in France is 10^{-6} mSv/a.

Non-radiological

Discharge of non-radiological liquid effluent from THORP reprocessing plant in the United Kingdom will produce pollutant concentrations less than 1% of relevant Environmental Quality Standards.

3.10.5. Decommissioning

In decommissioning reprocessing plants, due to the presence of contamination by fission products, residual plutonium and other alpha emitting nuclides, consideration has to be given to the production of aerosols and chemical vapours during decontamination, the production of metal particles from equipment dismantlement, and production of concrete dust during the demolition of structures.

3.10.6. Accident considerations

Fuel reprocessing plants have large inventories of radioactive materials, however, the operating characteristics of a reprocessing plant are intrinsically safe in that the temperatures and pressures used are close to or below ambient.

The solvent used was selected because of its high flash point (much higher than the operating temperature), thus reducing the risk of fire.

The plant design takes into account the potential for an explosion. Where an explosive atmosphere may exist, the equipment is designed to withstand the pressure-peak of the explosion. In addition, process instrumentation (including gas analysers) and design features (e.g. explosion-proof electrical equipment) are used to reduce the explosion risk.

Accidental release of activity from a highly active liquid fission product storage tank and a spent fuel storage pond due to loss of coolant is also considered of low probability. In the highly active storage tanks, the number of cooling components available is greater than that actually required so that failure of one or more cooling components would not constitute a failure of the system.

Criticality is prevented by using safe geometry for equipment, limiting the mass or concentration of fissile material at any point and using neutron poisons wherever required.

Accidental situations normally considered in the emergency planning process are exothermic chemical reactions, solvent fires, ignition of loaded ion-exchange resins, a criticality excursion, dropping

of irradiated fuel handling container and loss of cooling to high-activity liquid waste. Analysis shows that in a commercial plant of modern design, these situations do not lead to off-site consequences and allow sufficient time for mitigatory action to be taken.

3.11. WASTE MANAGEMENT

3.11.1. Basic process

The wastes considered in this section do not include tailings, which have been discussed in the section on milling (section 3.3). The section does not include the management or disposal of spent fuel as this is discussed in section 3.9.

The purpose of this section is to introduce the concept of waste management and waste conditioning as an integral part of the nuclear fuel cycle. It is recognized that the choice of waste disposal facility design, construction materials, treatment and conditioning methods will be dependent on the type of waste to be disposed of, and country and site specific. Therefore the reader is referred to the following publications for further detailed discussions of waste management practices, costs and impacts [8, 10, 15 and 16].

Keeping in mind that this is a general discussion, there is no attempt in the following section to reproduce this level of detail found in those publications.

Wastes produced by nuclear facilities are classified into categories based on several factors. Factors such as source of the waste, the half life of the radionuclides in the waste, the intensity of highly penetrating radiation, the condition of the waste i.e. liquid or solid, final disposal requirements, and the presence of non-radiological hazardous substances in the waste are used in waste classification. In the case of radioactive wastes containing non-radiological hazardous wastes, special care is taken to either reduce the non-radiological hazard in the process of origin or the waste is segregated and stored or disposed of separately.

Following the classification of wastes into categories, appropriate disposal options are proposed.

The decisions concerning the final classification and disposal options will rest with the national authorities.

The facilities of the nuclear fuel cycle other than mines and mills, and reactors produce small quantities of wastes that have to be sent to waste management facilities. The sources of raw wastes are shown in Fig. 12 for the once through fuel cycle and recycling option (MOX fabrication and reprocessing). Details on the quantities may be obtained in Refs.[10, 15]. However, it must be noted that current facilities are continually improving their waste management systems, and thus decreasing the amounts of waste generated.

The wastes arising from these facilities are usually treated and then conditioned prior to disposal by any number of means. Different countries use different methods. However, the treatment and conditioning may be done at the facility of origin or at a specially designed facility at the site of storage or disposal.

The treatment usually consists of some means of concentrating the contaminants by evaporation, ion exchange, physical compaction, incineration and chemical precipitation. The concentrate is then immobilized in a solid matrix, concrete, bitumen, glass, or synthetic polymers [10]. The purpose of the treatment and conditioning is to put the waste into a state that is suitable for storage, transportation and eventually disposal. The conditioned waste is also in a form that effectively inhibits the migration of the radionuclides.

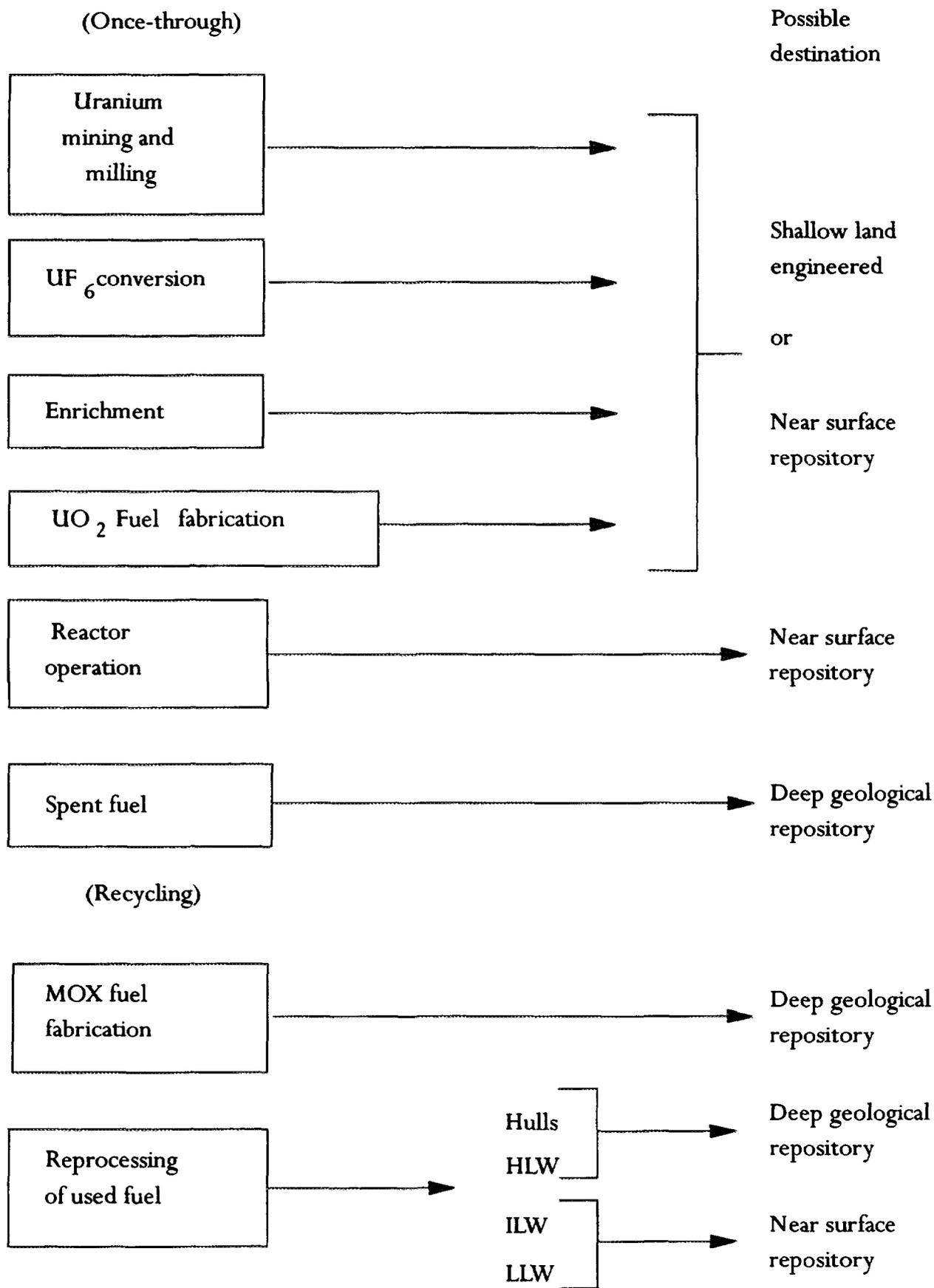


FIG. 12. Raw waste arising from the nuclear fuel cycle and possible destination

In some cases the wastes do not have to be conditioned either because:

- i) the amounts of radionuclides in them are so low that they may be released to conventional landfill with negligible impacts on either the environment and on humans; or
- ii) the material can be stored temporarily to allow the radionuclide content to reach the level in i) where it can be released without further control.

3.11.2. Resource requirements

Some land will be temporarily used for the buildings which house the handling, treatment and conditioning processes. This land will be reclaimed once the facility is decommissioned.

There will also be some land permanently taken by the disposal system. The size will be dependent on the system chosen and the amount of waste to be disposed of.

3.11.3. Releases

i) Incineration gases

Incineration is used as a means of reducing the volume of the waste. The process of incineration is carried out at elevated temperatures in order to ensure complete combustion of the waste. The gases from this process are vented to the atmosphere only after they have been passed through filters to remove particulates. The ash from incineration is passed back to the conditioning stream for immobilization.

The contents of the exhaust gases will be dependent on the materials incinerated and reflect the types of contaminants released by conventional incineration.

ii) Near surface disposal facilities

Near surface disposal facilities include trenches, covered mounds and sub-surface facilities excavated from 10 to 100 metres below the surface. The principle of waste isolation (from humans and the environment) is obtained in this case since:

- a) The matrix in which the waste has been placed is resistant to mobilization of the radionuclides, i.e. there are performance standards for the waste package.
- b) The design of the facility, the packaging of the waste and operating procedures are such that any migration from the facility is minimized.
- c) Administrative controls are in place until such time as the activity in the waste has decayed to acceptable levels. Such facilities are not used for the disposal of high level waste.

iii) Deep geological disposal facilities

These facilities are proposed to be constructed some tens to hundreds of metres below the surface in low permeability material. This kind of system is capable of isolating the waste for a lot longer than near surface facilities and thus can handle wastes that contain radionuclides with longer half lives, such as those found in high level wastes. The system was discussed in more detail in Section 3.9.

3.11.4. Potential health and environmental impacts during routine operation

i) Occupational

Radiological

The final stage of the disposal of solid wastes from the nuclear fuel cycle is not yet developed sufficiently to be able to make a detailed and separate examination of potential exposures. However,

UNSCEAR [6] states that some of the doses from the other parts of the fuel cycle are not expected to be significantly increased by the occupational exposures from waste disposal. This expectation is borne out by the data presented in UNSCEAR [6], where collective doses to workers in waste processing at LWRs in the USA represent a small fraction of the total collective dose among all workers at these facilities.

Non-radiological

Wastes originating from nuclear fuel cycle facilities are segregated, conditioned and packaged. This, along with appropriate procedures, reduces worker exposure to conventional hazards.

ii) *Environmental*

Radiological

Potentially the principal process for the release of radionuclides is slow transport by groundwater. Site location and design will be chosen to minimize the risk of radionuclide release over sufficiently long periods to allow the fission products which pose the highest risk to humans, to decay.

Modelling studies carried out using the foregoing assumptions, indicate that the doses to humans would be very small [10]. The principles of radiological isolation and the use of multiple barriers would also reduce any possible conventional impacts.

Non-radiological

No significant amounts of effluent are expected to result from repositories.

3.11.5. Decommissioning

A repository will be shut down when it is completely filled to its licensed quantity of radioactive waste. Shutdown is followed by a period of surveillance during which access to the site is restricted. The most important steps in shut down are backfilling or stowing of remaining space, sealing of access tunnels or shafts, and decommissioning and removal of surface buildings and installations.

As part of the mitigative measures, records of waste inventories and relevant site construction and operating data should be kept in suitable archives for an appropriate period of time. It may also be necessary to impose restrictions on the use of the land surface above the repository and in the vicinity of the surface facilities for a certain period of time after the repository is shut down and sealed.

3.11.6. Accident considerations

Appropriate consideration must be given both to discrete events and to physical processes, such as those caused by human activities (alterations of biosphere and hydrology, mining and drilling activities), hydrological and geological natural processes (groundwater flow, erosion, faulting/seismicity, uplift and volcanism), and waste emplacement and repository construction (thermal, chemical, mechanical and radiological effects). The consequence of many events and processes can be reduced by careful site investigation and selection processes. Provision must be made to guard against human intrusion. For shallow-ground repositories, this can be controlled by institutional means. However for deep geological disposal other means must be used. Details of these considerations may be found in other IAEA publications [10].

4. CONCLUDING REMARKS

Comparative assessment of health and environmental impacts of different electricity generation options is essential for planning and implementing sustainable strategies for electricity service supply. This report describes in a very general way the health and environmental aspects of the nuclear fuel cycle. The releases have not been quantified as this will be available from the DECADES database. Relevant references are given to aid the reader in discovering more about the fuel cycle in general, or about a facility in particular.

The list of radioactive and non-radioactive emissions considered at each stage of the nuclear fuel cycle may not be exhaustive, but represent the major releases from the facilities. In addition, substances that are hazardous have also been identified. Where impacts on health or environment are quoted, they are intended to be illustrative and are based on previous studies by other organizations such as UNSCEAR. In discussing impacts to humans, use has been made of the concept of collective dose. Further details of this concept may be obtained from UNSCEAR [6].

While reviewing and compiling available information, the experts pointed out the need for establishing a comprehensive and harmonized data base and accounting framework for all emissions arising from different electricity generation strategies. The database should be constructed in such a way that meaningful comparisons can be carried out.

Potential impacts on human health and the environment of radioactive and non-radioactive emissions have been studied and evaluated thoroughly. However, there is a need for a more comprehensive approach to such emissions from other electricity generation strategies.

Specific topics requiring further research and analysis include:

- harmonization of risk assessment between radionuclides and conventional chemicals;
- establishment of harmonized (radiological/non-radiological) indices of environmental harm, and quality;
- comprehensive review of the accidental risks associated with different facilities and electricity generation options;
- assessment of the environmental impacts of atmospheric emissions such as CO₂.

The report is limited to chosen health and environmental aspects of the nuclear fuel cycle facilities, namely to evaluation of collective and in some cases individual average doses and radioactive releases from the facilities. It also does not consider non-radiological hazards to the workers and the public, so that non-radiological health effects connected with the construction, operation and decommissioning of facilities are not discussed.

The reader looking for guidance as to the structure of a comprehensive health and environmental impact analysis, which should cover all stages of the nuclear fuel cycle and all kinds of health and environmental impacts, is referred to other IAEA documents.

APPENDIX A

EXAMPLE OF AN EVALUATION OF OPERATIONAL IMPACTS AT A URANIUM MINE AND MILLING FACILITY

1. Summary of Impacts in Uranium Mining and Milling

Some of the various impacts that may result from uranium mining and milling are limited to certain types of operations. The following table lists the various impacts and shows the types of operations in which they are likely to occur.

The letters appearing in the column labelled "Operation" indicate where the impacts may occur, and have the following meanings:

- O = open pit mining
- U = underground mining
- A = in situ leach using acid
- B = in situ leach using alkaline
- M = milling (including tailings management)

The various agents are assigned a code which is given in the column labelled "ID" for reference later in this document.

Hazard	Pathway	Group at risk	Agent	Operation	ID
Radiological	Air	Workers	External Radiation	O U M	R1
			Radon Progeny	O U A B M	R2
			Skin Dose	O U M	R3
			Ingestion	O U M	R4
			Long lived dust	O U M	R5
	Others	Radionuclide Release	O U A B M	R6	
	Water	Others	Radionuclide Release	O U A B M	R7
Toxicological	Air	Worker	CO and CO ₂	O U A B M	T1
			NO, NO _x	O U	T2

Hazard	Pathway	Group at risk	Agent	Operation	ID
Toxicological	Air	Worker	Diesel particulates	O U	T3
			Blasting fumes	O U	T4
			Noise	O U M	T5
			Temperature/humidity	O U A B M	T6
	Water	Others	Metals leaching	O U A B M	T7
Other	n/a	Worker	Blasting	O U	O1
			Travel/Transportation	O U A B M	O2
			Ground failure (major and minor)	O U	O3
			Fire	O U M	O4
			Vibration	U	O5
			Damp conditions	U	O6
			Heavy equipment	O U A B M	O7
			Reagents	A B M	O8
		Others	Ground opening	O U	O9
			Dam failure	M	O10
			Land alienation - temporary	O U A B M	O11
			Land alienation - permanent	O M	O12
Other	n/a	Others	Subsidence	U	O13

2. Evaluation of Impacts

Uranium mining and milling can impact workers, the local population and the general environment. Such impacts can be of three types: radiological, toxicological and other (see section 1). The significance of these impacts for a particular mine or mill is controlled, primarily, by four factors: the type of operation, the grade of ore being mined or milled, the age of the facility (since older facilities do not, in general, have the same radiation and environmental protection controls available), and the density of the local population. Combining the factors leads to the possibility of 40 different categories of facilities that may exist. In practice, however, not all factors are significant in all cases, and the number of cases is less.

It is impossible to develop a single model that is representative of all categories, as there are major differences that cannot be averaged when determining the impacts. However, it is theoretically possible to develop a model for each category, and then to determine the total impact of uranium mining and milling operations worldwide by summing the impacts category, weighted by the amount of uranium that comes from the category.

To make this task manageable, the following table has been developed. It identifies the impacts that **may** be significant under a particular set of circumstances. In order to determine the total impact, only those impacts that are significant need be evaluated. While other impacts may be present, their contribution to the total impact is likely to be small.

The assumption is made that appropriate management practices are used to limit certain impacts and, therefore, such impacts are not included. Should this not be the case, it will be necessary to consider all impacts that may result (see section 1).

It should be noted that the impacts on the local population and on the environment are likely to be different during operations and after closure of the facility. The entries under milling include the possible impacts from the tailings management facility during operations and after closure.

The entries for in situ leaching include the possible impacts from both the mining and processing. For conventional mining and milling operations, it will generally be necessary to consider the impacts from mining and milling independently.

The letters and numbers appearing in the columns labelled "Significant Impacts" indicate the identification of the impact that may occur, as listed in the column marked "ID" in the table in section 1. Where there is no entry, no significant impacts are anticipated.

It is not intended that the differentiation between high and low grade, between a new facility and an old one, and between dense population and sparse population in the vicinity of the facility be absolute. As a general guide in interpreting the table, the following definitions may be used:

High grade:	uranium content of the ore greater than 0.5%:
Low grade:	uranium content of the ore less than 0.5%

New facility: facility constructed to currently accepted environmental and safety standards
 Old facility: other facilities
 Dense population: an average of greater than 3 people per square kilometer living within 25 kilometers
 Sparse population: an average of less than 3 people per square kilometer living within 25 kilometers

Operation				Significant Impacts				
Type	Grade of ore	Age of facility	Local Population	Workers	Population		Environment	
				During	During	After	During	After
Open pit	High	New	Dense	R1,2,5 T2,4,5,6 O1,2,3,7	R6 O11	R7 T7 O9	R6,7 T7	T7
			Sparse			O9		
		Old	Dense	R1,2,5 T2,4,5,6 O1,2,3,7	R6 T7 O11	R7 T7 O9	R6,7 T7	R7 T7
			Sparse		R6 T7	O9		
	Low	New	Dense	T2,4,5,6 O1,2,3,7	R6 T7 O11	T7 O9	T7	T7
			Sparse			O9		
		Old	Dense	T2,4,5,6 O1,2,3,7	R6 O11	T7 O9	R7 T7	R7 T7
			Sparse			O9		
Under-ground	High	New	Dense	R1,2,3,5 T2,3,4,5 O1,2,3,4,5,6,7	R6 O11	O13	R6	
			Sparse					

Operation				Significant Impacts				
Type	Grade of ore	Age of facility	Local Population	Workers	Population		Environment	
				During	During	After	During	After
Underground	High	Old	Dense	R1,2,3,5 T2,3,4,5 O1,2,3,4,5,6,7	R6 O11	O13	R6	R7
			Sparse					
	Low	New	Dense	R1,2,5 T2,3,4,5 O1,2,3,4,5,6,7	R6 O11	O13		
			Sparse					
		Old	Dense	R1,2,5 T2,3,4,5 O1,2,3,4,5,6,7	R6 O11	O13	R6	
			Sparse					
In situ leaching - acid	High or low	New or old	Dense or sparse	T6 O2,7,8	O11	R7 T7		R7 T7
In situ leaching - alkaline	High or low	New or old	Dense or sparse	T6 O2,7	O11			
Milling	High	New	Dense	R1,2,3,4,5 T5,6 O2,4,7,8	R6,7 O11	O10,12	R6,7	O10
			Sparse			O12		
		Old	Dense	R1,2,3,4,5 T5,6 O2,4,7,8	R6,7 O11	R7 T7 O11	R6,7	R7 T7 O10
			Sparse			R7 T7 O12		
	Low	New	Dense	R1,2,4,5 T5,6 O2,4,7,8	O11	O12		O10
			Sparse			O12		

Operation				Significant Impacts				
Type	Grade of ore	Age of facility	Local Population	Workers	Population		Environment	
				During	During	After	During	After
Milling	Low	Old	Dense	R1,2,4,5 T5,6 O2,4,7,8	R6,7 O11	O12	R7	R7 T7 O10
		Old	Sparse			O12		

3. An Example

Mine: Key Lake, Canada
 Type: Open pit and mill
 Grade of ore: High
 Age of facility: New (1983)
 Local population: Sparse
 Production: 5314 tonnes of uranium in 1993 (16.3% of world production)

Possible significant impacts of the mine

- R1 - External radiation exposure of workers
- R5 - Long lived radioactive dust exposure of workers
- R6 - Impact on the environment of radionuclide releases to the air during operations
- R7 - Impact on the environment of radionuclide releases to the aquatic environment during operations
- T2 - NO, NO_x exposure of workers
- T4 - Blasting fume exposure of workers
- T5 - Noise exposure of workers
- T6 - Temperature and humidity exposure of workers
- T7 - Impact on the environment of metals leaching into the aquatic environment
 - During operations
 - After the completion of operations

- O1 - Blasting accidents to workers
- Of - Travel and transportation accidents to workers
- O3 - Ground failure accidents to workers
- O7 - Heavy equipment accidents to workers
- O9 - Accident risk resulting from the existence of the open pit after completion of operations

Possible significant impacts of the mill

- R1 - External radiation exposure of workers
- R2 - Radon progeny exposure of workers
- R3 - Radiation skin dose of workers
- R4 - Ingestion of radioactive material
- R5 - Long lived dust exposure of workers
- R6 - Impact on the environment of radionuclide releases to the air during operations
- R7 - Impact on the environment of radionuclide releases to the aquatic environment during operations
- T5 - Noise exposure of worker
- T6 - Temperature and humidity impact on workers
- Of - Travel and transportation accidents to workers
- O4 - Fire occurrence
- O7 - Heavy equipment accidents to workers
- O8 - Accidents to workers involving reagents
- O10 - Probability and environmental consequences of a dam failure after completion of operations
- O12 - Amount of land that will be alienated from future use

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GLOSSARY

alpha bearing waste. Waste containing one or more alpha emitting radionuclides, usually actinides, in quantities above acceptable limits for uncontrolled release. The limits are established by the national regulatory body.

criticality (accident). A criticality accident occurs when fissile materials accumulate in an amount and configuration which allow a chain reaction to be sustained uncontrollably. Its consequences are a sudden increase of the neutron flux and a corresponding energy release.

collective dose. This quantity takes account of the number of people exposed to a source by multiplying the average dose to the exposed group from the source by the number of individuals in the group. If several groups are involved, the total collective quantity is the sum of the collective quantities for each group. The unit of these collective quantities is the man sievert.

critical group. For a given radiation source, the members of the public whose exposure is reasonably homogeneous and is typical of individuals receiving the highest effective dose from the source.

effective dose. The amount of absorbed radiation per unit mass of matter, it is expressed numerically in sieverts, symbol Sv, as the unit of effective dose. 1 Sv=100rem.

light water reactor. A reactor in which the chain reaction is sustained primarily by fission brought about by thermal neutrons, i.e. neutrons which are in thermal equilibrium with the material in which they are moving. Such reactors use light water as a moderator to slow down the neutrons produced in fission to thermal energies.

heavy water reactor. A reactor in which the chain reaction is sustained primarily by fission brought about by thermal neutrons, i.e. neutrons which are in thermal equilibrium with the material in which they are moving. Such reactors use heavy water as a moderator to slow down the neutrons produced in fission to thermal energies.

pathway analysis. The analysis is conducted by a pathways model. Pathways model is a mathematical description, usually in the form of a computer algorithm, that determines the relative significance of possible radionuclide transport vectors, e.g. air, ground water, surface water, intrusive roots, animals, etc.

recommended dose limits (ICRP). The radiation exposure limits is recommended by the International Commission on Radiological Protection (ICRP). The dose limits of effective dose are 1mSv in a year for public and 20 mSv per year for occupational. Refer to ICRP publication 60 in detail.

Sources: IAEA-TECDOC-447 Radioactive Waste Management Glossary, second edition 1988.
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PART II
SITE REPORTS

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EVALUATION OF ENVIRONMENTAL IMPACTS OF URANIUM MINING AND MILLING OPERATIONS IN SPAIN

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Abstract

Uranium mining and production activities have been carried out by ENUSA since 1973. This report describes the evaluation of environmental aspects connected with uranium mining and milling.

1. INTRODUCTION

The Empresa Nacional del Uranio, S.A. (ENUSA), is a Spanish company created in 1972. ENUSA provides products and services related to the front end of the nuclear fuel cycle.

Uranium mining and production activities have been carried out by ENUSA since 1973 in the area of Ciudad Rodrigo (Fig. 1), province of Salamanca, they were based on open pit mining, heap leaching and a hydrometallurgical plant (plant ELEFANTE) for obtaining uranium concentrates from the pregnant liquids. During 1993 the plant ELEFANTE was stopped and a new plant QUERCUS was started with dynamic leaching. The nominal capacity of the new plant is 950 t U_3O_8 /year, nowadays because of the low price of uranium, the facility is running at a production rate of 300 t U_3O_8 /year.

ENUSA has incorporated in the design, construction and operation of the QUERCUS plant, the best available technology to reduce the environmental impact, keeping in mind the experience gained during the operation of uranium facilities over the past 25 years.

The purpose of this article is the evaluation of the environmental impact produced in the operation of the new plant.

2. DESCRIPTION OF THE FACILITIES

2.1 Open Pit Mining of the Fe and De deposits

The Fe and De deposits are an anarchical mineralization in the fissures and faults of the Cambrian Slates (stockwork type), whose shallow overburden allows open pit mining. Based on a cut-off grade of 280 ppm of U_3O_8 , we can calculate the ore reserves of this deposit to be some 10,000 tons of U_3O_8 . The average grade is 700 g U_3O_8 /t.

The mine benches are 6 meters high. Blasting is done carefully by minimizing rock movements to avoid mixing ores and wastes. By means of a radiation monitor, the rocks are classified as ores or wastes, wastes are transported to a waste rock disposal site located near the mine, while ores are taken to the plant for milling operations.

The lay-out of both the Uranium Mine and the QUERCUS Plant is shown in Fig. 2.



FIG. 1. Uranium Facilities in Spain

2.2 Uranium Mill

The flow diagram (Fig. 3) of the QUERCUS plant is as follows:

The ore is crushed to a size less than 100 mm and it is either placed in temporary storage or fed directly to the classification section of the mill. The ore is classified in three different groups according to its size: less than 1 mm, between 1 mm and 10 mm and more than 10 mm. The ore with a size of less than 1 mm contains most of the uranium and it is tried in the dynamic leaching plant. The remaining part of the mineral is treated by static leaching (size between 1 and 10 mm) or taken to the waste rock disposal (size larger than 10 mm).

The fine slurry is taken to the Acid Leach Circuit where it is subjected to the attack of hot dilute sulfuric acid. Liquid and solids are then separated in Counter Current Decantation where five continuous thickeners are used to wash the solids free of dissolved uranium. A pregnant solution overflows to the Classification Circuit of 0,5 to 0,6 g of U_3O_8 per litre, is produced. The solution discarded with the solids to the tailings dam allows a bleed of impurities from the plant.

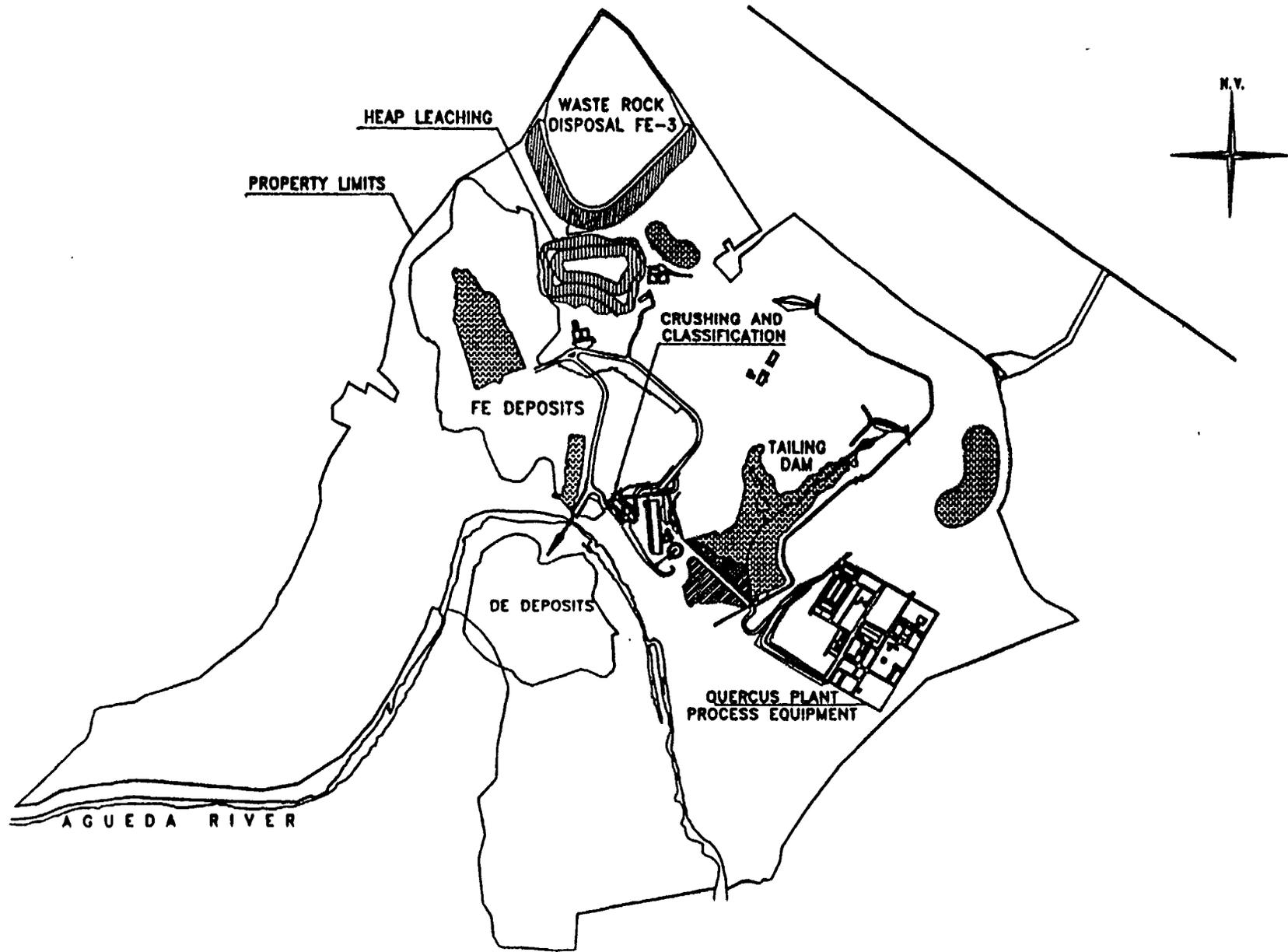


FIG. 2. *Uranium Mine and Plant Quercus Lay-out of Facilities*

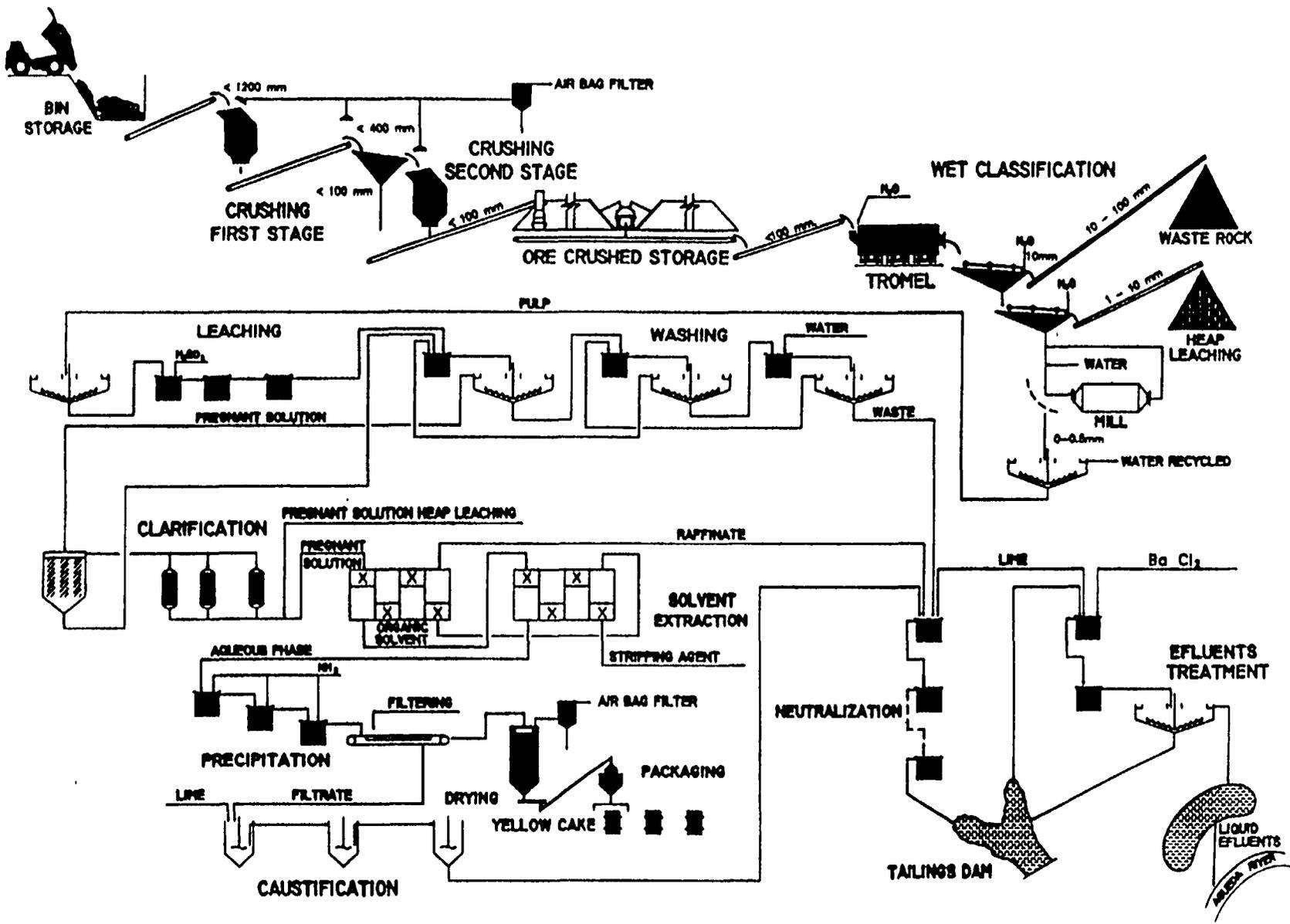


FIG. 4. Layout of Quercus Plant

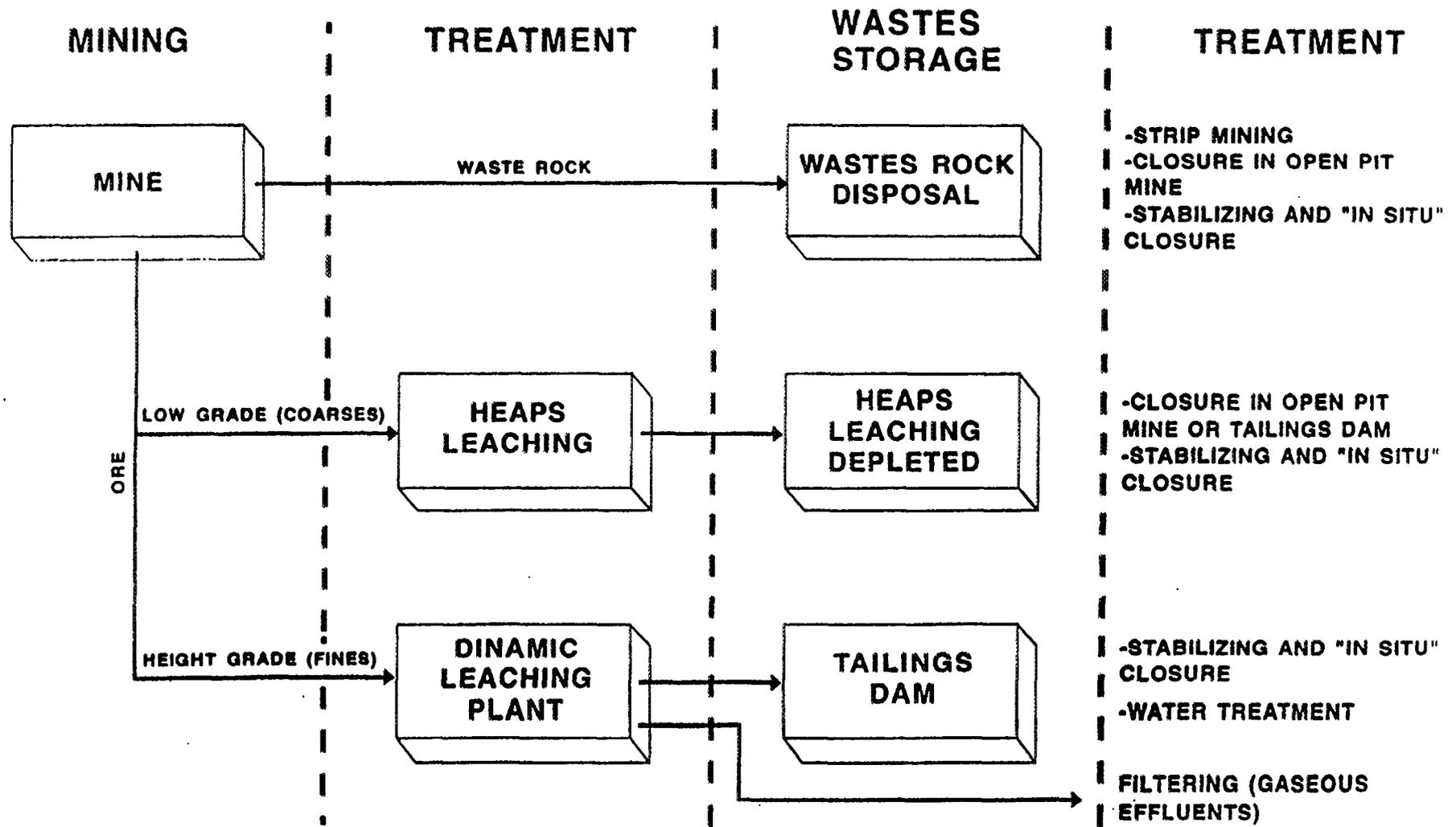


FIG. 5. Mangement of Wastes in Uranium Facilities

The Drying Section has a spray drying plant with rotary atomizer operating at 400°C. From the drier, the solids go into a storage bin for automatic packaging in 200 l steel drums for shipment. Overall recovery is more than 90% of the uranium in the ore into a product containing more than 90% U₃O₈.

The layout of the plant is shown in Fig. 4.

3. MANAGEMENT OF WASTES IN URANIUM FACILITIES

At present, in ENUSA's mining and milling operation, the different types of wastes and its treatment to mitigate the environmental effects are indicated in Fig. 5.

3.1 Mining Wastes

This is waste rock which is stored at waste rock deposits located near the mine. The average uranium concentration of this waste material is less than 100 ppm.

The granulometry is determined by the system employed to remove the rock. Since ENUSA uses blasting techniques, the granulometry is very broad (< 1.000 mm), a factor which has a favourable effect on the environmental stability of the deposited waste.

The volume of this material varies according to the ore cut-off grade and the waste/ore ratio. The annual average is 3×10^6 tons per year.

Table I shows the chemical composition of the ore, as well as the extreme values what stands the abundance of iron, alumina, and silica, due to the siliceous nature of the rock and the presence of clay.

Table I. Quantitative Chemical Analysis of the Most Important Elements (%) Contained in the Ore

DETERMINE	AVERAGE VALUES	EXTREME VALUES
Al ₂ O ₃	20,0	15,0 - 27,0
CaO	0,4	0,1 - 1,0
Fe ₂ O ₃	7,0	5,0 - 8,5
SiO ₂	65,0	55,0 - 80,0
CO ₃	0,1	0,01 - 0,5
SULFIDE (as SO ₄)	0,2	0,05 - 0,5
TOTAL S (as SO ₄)	1,4	0,1 - 6,0
CALCINATION loss	5,0	3,0 - 7,0

3.2 Milling Wastes

a) Solid and liquid wastes

The mill operation generates both radioactive and non-radioactive wastes. The tailings represent the bulk of both radioactive and non-radioactive wastes. With the exception of the recovered uranium and some process losses, tailings account for practically all of the ore solids and the process additives, including water.

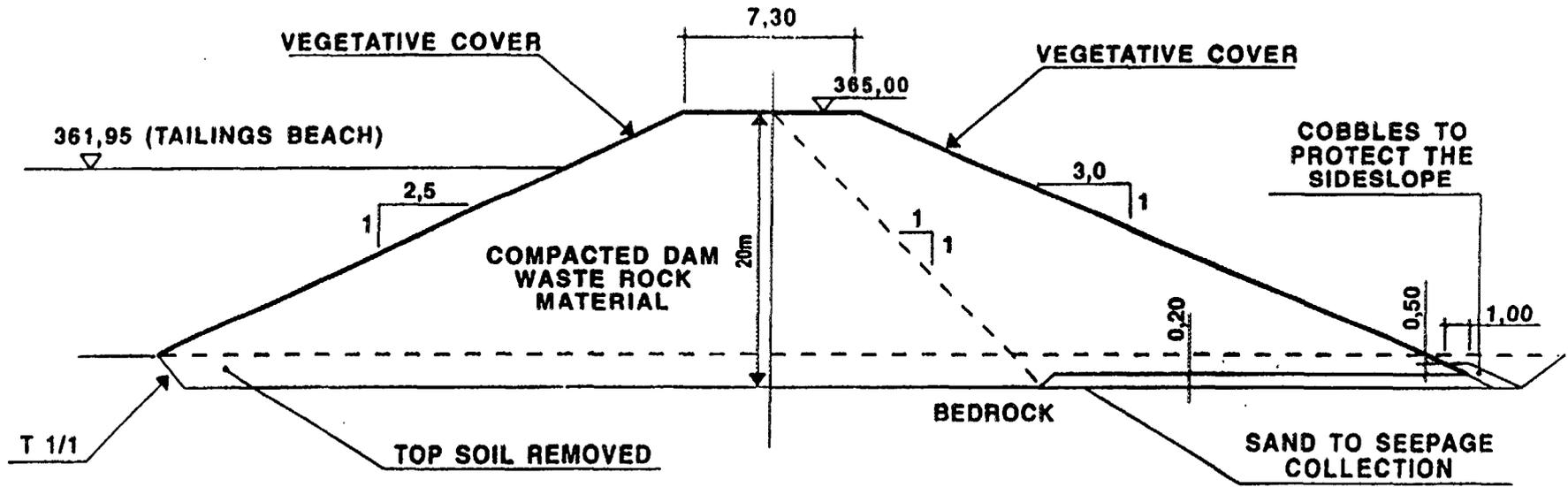


FIG. 6. Tailings Dam Section

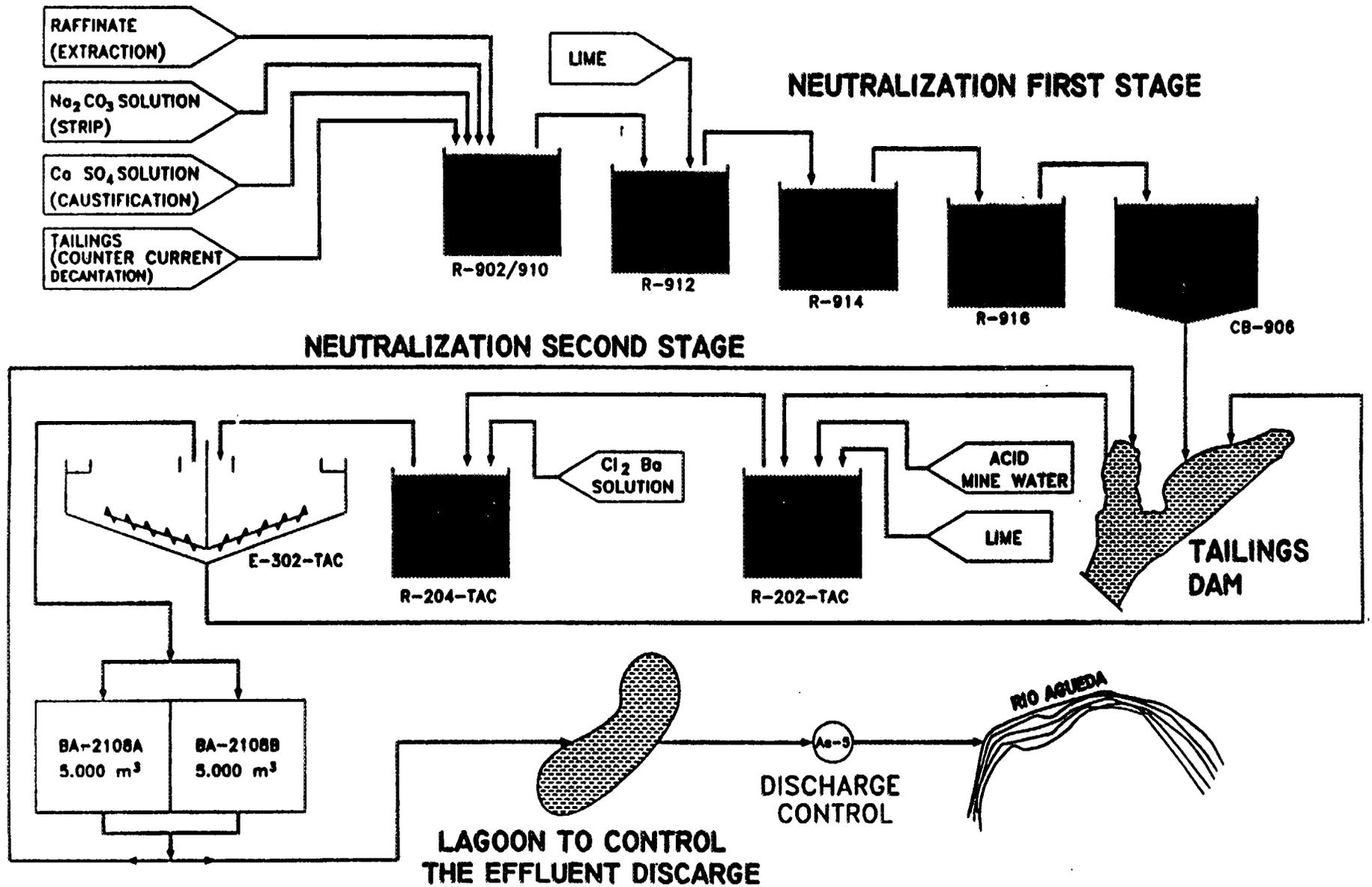


FIG. 7. Liquid Effluent Treatment and Discharge System

The slurried tailing material is pumped from the mill through plastic pipes to a tailings dam. The tailings dam (Fig. 6) was built according to the Regulatory Guide 3.11 of the Nuclear Regulatory Commission (U.S.A) and the National codes; its main characteristics are as follows:

- Cell area $20 \times 10^4 \text{ m}^2$
- Capacity $2,2 \times 10^6 \text{ m}^3$
- Height 50 m
- Side slope inclination 3/1 and 2,5/1

At the present production rate the mill generates about 70.000 MT of dry tailings slurried in water to about 30% solids by weight.

The effluent is neutralized before it gets to the tailings dam. The system for the treatment of the liquid effluent is shown in Fig. 7.

The limits to be accomplished for the liquid effluent, after treatment and before being discharged to the river, are indicated in Table II.

Table II Maximum Permissible Concentration of Radionuclides and Chemicals in Liquid Effluent

CHEMICALS

CHEMICAL (mg/l)	LIMITS R.D.P.M
PH	5,5-9,5
SUSPENDED SOLIDS	150
Al	1
Cd	0,1
Fe	2
Mn	2
Ni	2
Pb	0,2
Cu	0,2
Zn	3
Cl-	2000
S04=	2000
NH3	15

RADIOACTIVE

- ANUAL ACTIVITY DISCHARGED Ra-226 : $1,65 \times 10^9 \text{ Bq}$
- MAXIMUM INCREASE IN THE RIVER WATER ACTIVITY Ra-226 : $3,75 \text{ Bq/m}^3$
- TOTAL ALPHA ACTIVITY IN THE RIVER (LIMIT OF ENUSA PROPERTY) : 555 Bq/m^3

b) Airborne effluent

The major sources of both radioactive and non-radioactive gaseous effluent are indicated in Table III. This effluent is filtered by means of air bag filters before its release to the atmosphere.

The limits to be accomplished for the airborne effluent, after treatment and before being discharged to the atmosphere, are indicated in Table III.

Table III Limits for Airborne Effluent

EMISSION SOURCE	AIR VOLUMEN m ³ /h	LIMITS OF SOLIDS CONCENTRATION mg/Nm ³
ORE CRUSHING	150000	< 15
PRODUCT DRYING	4500	< 5
YELLOW CAKE PACKAGING	10500	< 5

4. CLAIMED INNOVATION IN URANTUM MINES AND MILLING WASTES

The major option in the long-term management of the tailings is that they are confined in an impoundment using the best available technology. According to ENUSA's experience, it appears that the tailing management is first of all a water management problem. The release of tailings effluent to the environment has to be focussed on the reduction of the effluent volume that has to be treated in order to be discharged without damage. This effluent is characterized by:

- acidity related to the sulfide oxidation
- high heavy metals toxic elements and radionuclides content
- high dissolved salt content such as chlorure, sulfate,

For this purpose, it is very important to proceed step by step to reach a better understanding of the distribution of radionuclides and toxic elements in the tailings and their solubization and migration properties.

Related with the above, at the present ENUSA is working in the following projects:

- Detailed knowledge of the mineralogy and geochemistry of mill tailings
- Development of new analytical techniques for location of the radionuclides in the tailings
- Location of radionuclides and heavy metals in mill tailings, sludges and mine wastes
- Microfiltration and ultrafiltration of waters to reduce the sludges volume
- Increasing the size of precipitated crystals in the sludge
- Optimizing the acid waters treatment
- Development of enhanced leaching techniques of mining waste for faster decommissioning of the sites.

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PRACTICAL THRESHOLD LIMIT VALUES IN A CONVERSION PLANT

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Abstract

Comurhex is a subsidiary of the Cogema group, and its business is the conversion to hexafluoride of the uranium contained in a variety of mining concentrates of differing origins. This report gives an example of the evaluation of the health aspects of a conversion plant.

INTRODUCTION

The company has two production units: its **Malvési Works**, which produces uranium tetrafluoride, UF_4 , and its **Pierrelatte Works**, where the tetrafluoride is converted to the hexafluoride, UF_6 .

The end-product, the hexafluoride, subsequently goes to enrichment plants in which the proportion of the uranium-235 isotope is stepped-up by the gaseous diffusion or centrifuging processes.

MALVESI

The process and-the installations operated at Comurhex's Malvési Works are illustrated diagrammatically in Figure 1.

1. Digestion

The incoming ore concentrates (containing some 70 - 75% uranium) are digested in concentrated nitric acid. After a "maturing" period designed to assist the flocculation of insolubles (silica et al), if any, the liquor so obtained, containing some 650 grams of uranium per litre is filtered through a set of three back-washed rotary vacuum filters precoated with diatomaceous earth.

This yields a filtrate consisting of an impure solution of uranyl nitrate containing 450g uranium per litre (450g U/l). The insolubles filtered off, still containing traces of uranium, go to the Recovery plant (cf. 5 below) and thence to the waste lagoons.

2. Purification

The uranyl nitrate solution leaving the filters is purified by a two-stage liquid/liquid extraction process. In the first stage, the aqueous uranyl nitrate solution is intimately mixed, in a stirred column, with a solution of tributylphosphate (TBP) in dodecane. At the interfaces between the droplets of aqueous and organic phases the uranyl nitrate and TBP react to form a complex which passes into the dodecane phase.

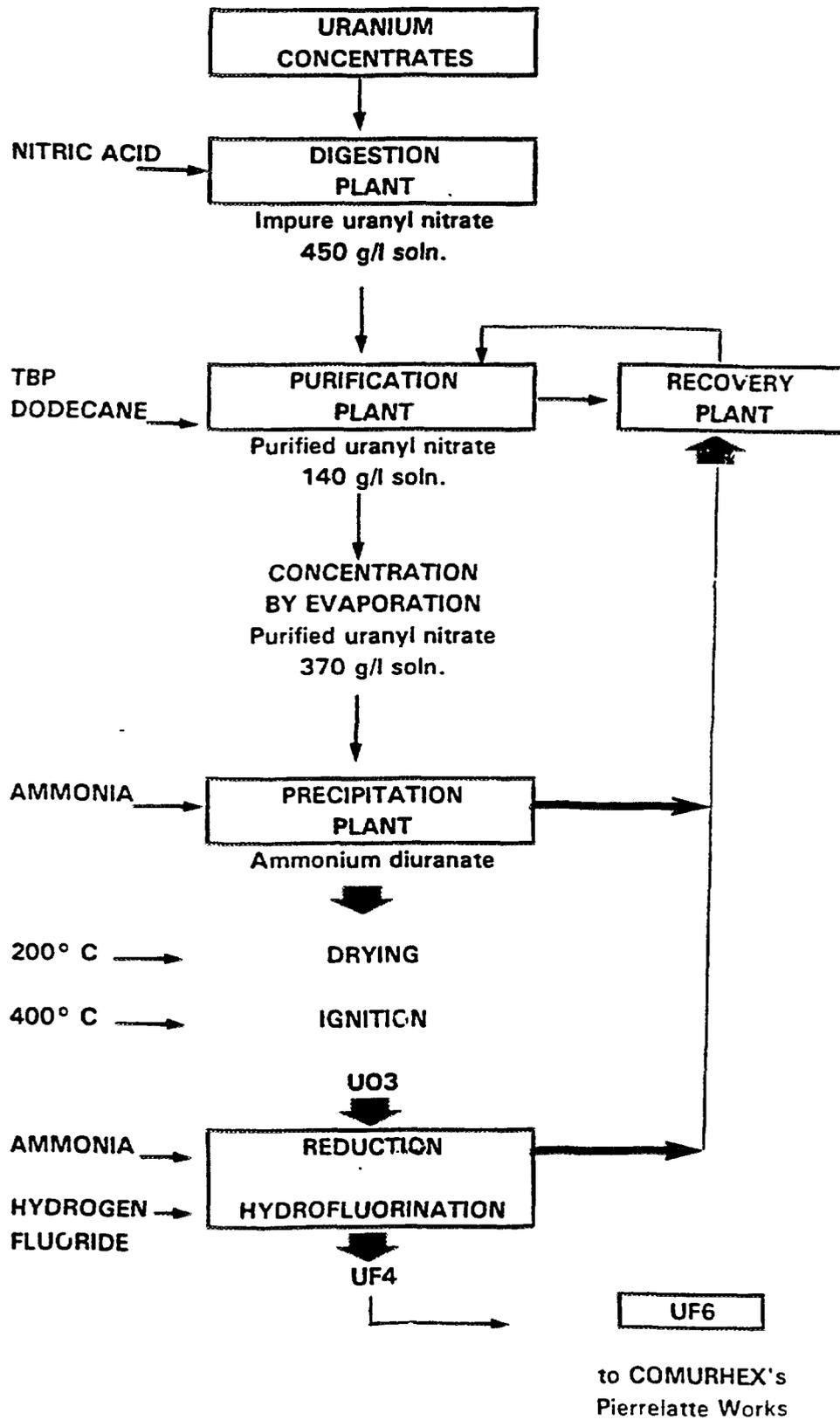


FIG. 1. Process and installations operated at the Comurhex Malvési Works

The organic phase, now loaded with uranium, is washed and the uranyl nitrate finally taken back into aqueous solution with demineralised water in a re-extraction column.

The solvent freed of uranium is regenerated and recycled to the extraction stage, while impurities, which contain less than 10 mg U/l, go, via the Recovery plant and after neutralization, to the lagoons.

The pure uranium nitrate solution, leaving the purification plant at a strength of 135 - 140g U/l, now goes to a four-stage steam-heated evaporator in which it is concentrated to a final 370g U/litre.

3. Precipitation and ignition

The next stage is to treat the uranyl nitrate solution with gaseous ammonia to precipitate the uranium as the diuranate.

The slurry so obtained is dewatered by rotary vacuum filtration and the ammonium nitrate mother liquor analysed and subsequently reused.

The ammonium diuranate (ADU) filter-cake is conveyed directly to calciners in which it is first dried, then ignited at some 400°C to obtain uranium trioxide, UO₃.

4. Hydrofluorination

The orange-coloured trioxide now goes to the Hydrofluorination plant, where it is first reduced to the brown dioxide, UO₂, and then reacted with hydrogen fluoride, HF, to obtain the tetrafluoride.

Both these operations are carried out in one and the same moving-bed furnace, in which the bed of solid material flows downwards. Gaseous ammonia injected into the upper part of the furnace is thermally cracked by heating to 700°C; the hydrogen so produced reduces the uranium trioxide to the dioxide and the dioxide finally reacts with hydrogen fluoride injected at the base of the furnace to yield the green tetrafluoride.

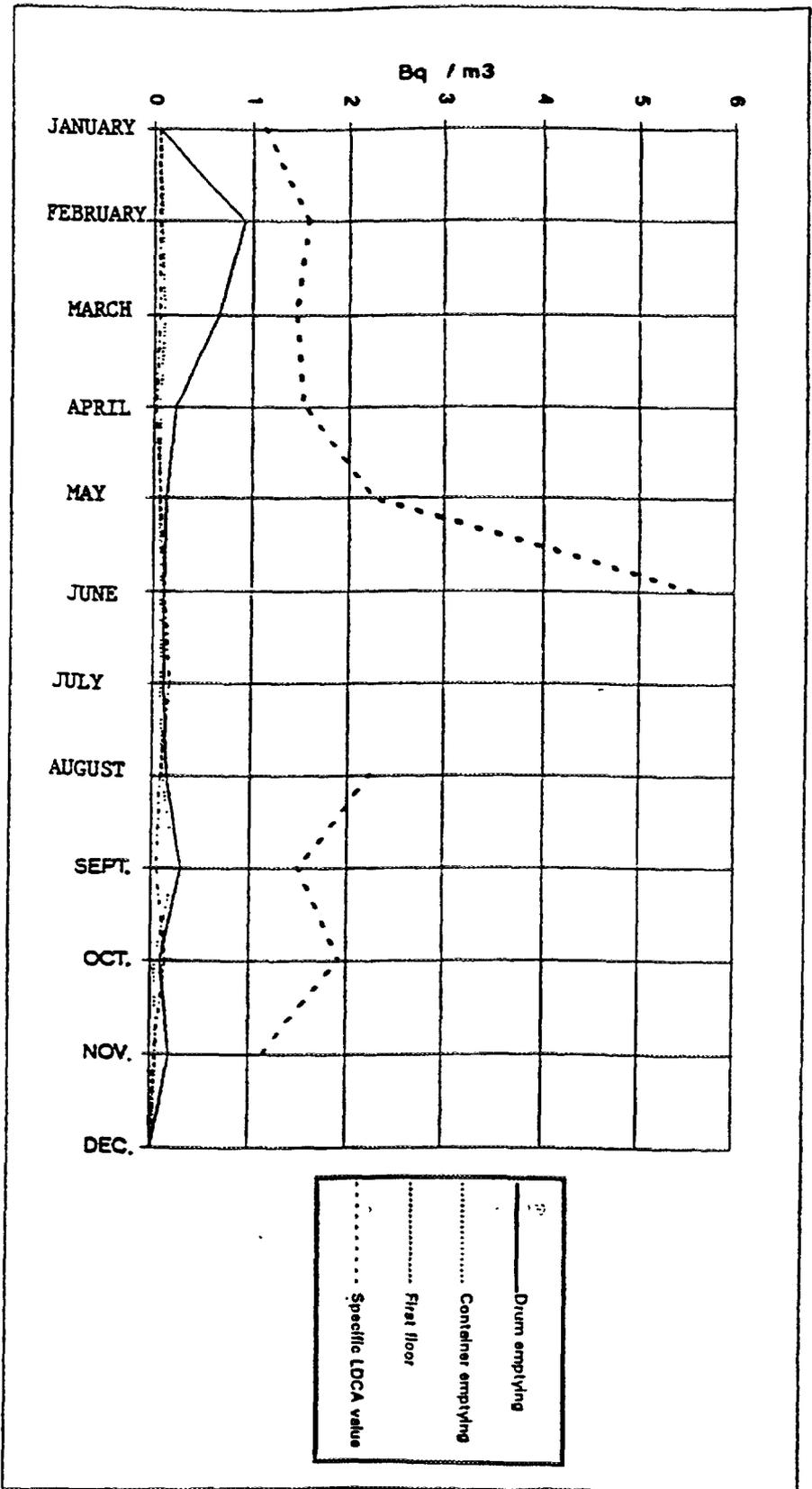
The uranium tetrafluoride leaving the furnace goes to a storage bin, from which it is loaded into road-tankers for delivery to Comurhex's Pierrelatte Works.

5. Recovery

All uranium-containing by-products of this conversion process are taken from the various production plants to a Recovery plant in which they are processed for complete recovery of uranium contents.

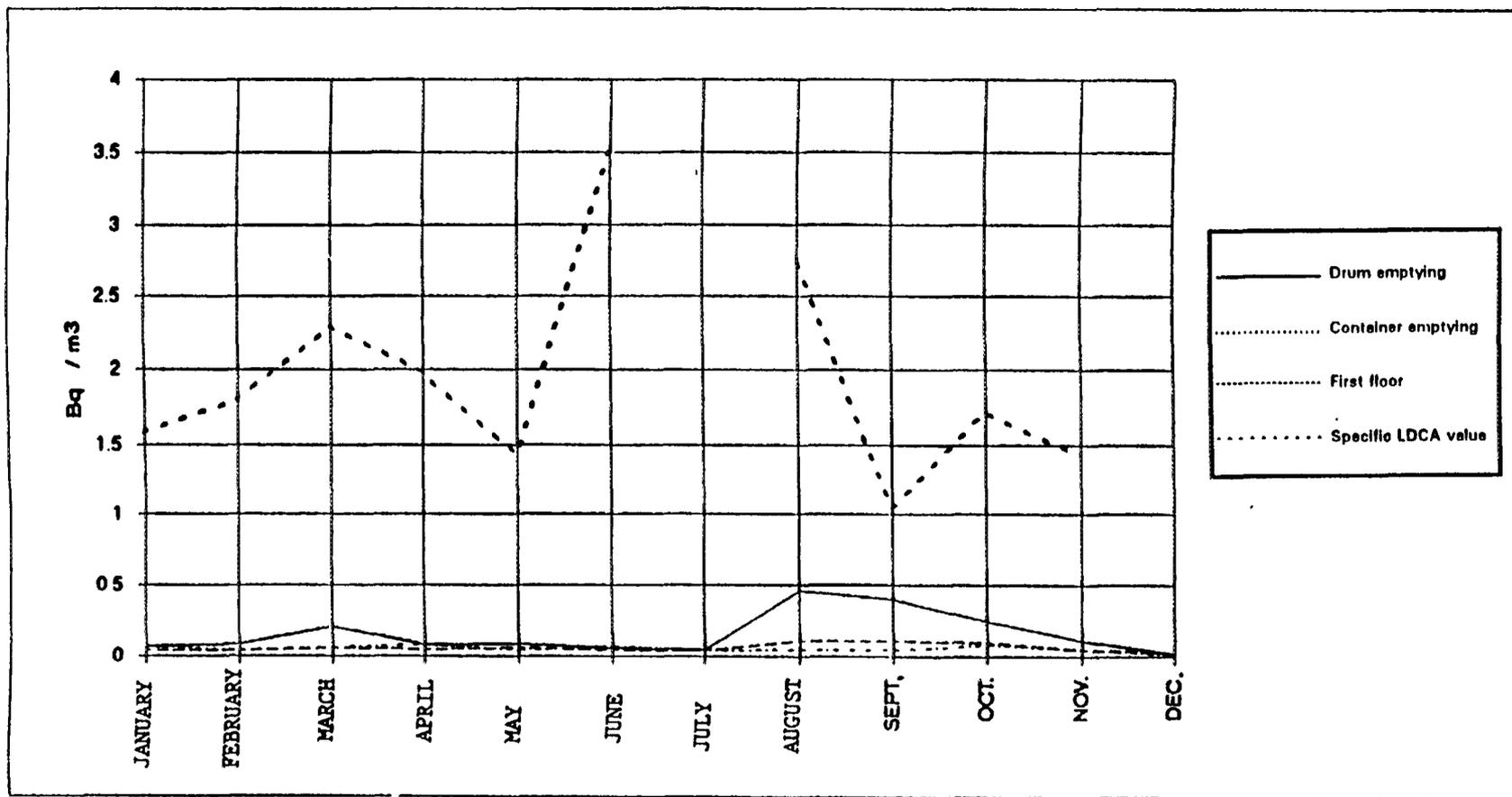
All the uranium thus recovered is recycled to the Purification stage. Residual uranium free liquors are neutralized with lime before going to the lagoons.

The Recovery plant also processes solid residues originating from Comurhex's Pierrelatte Works and from a number of COGEMA plants.



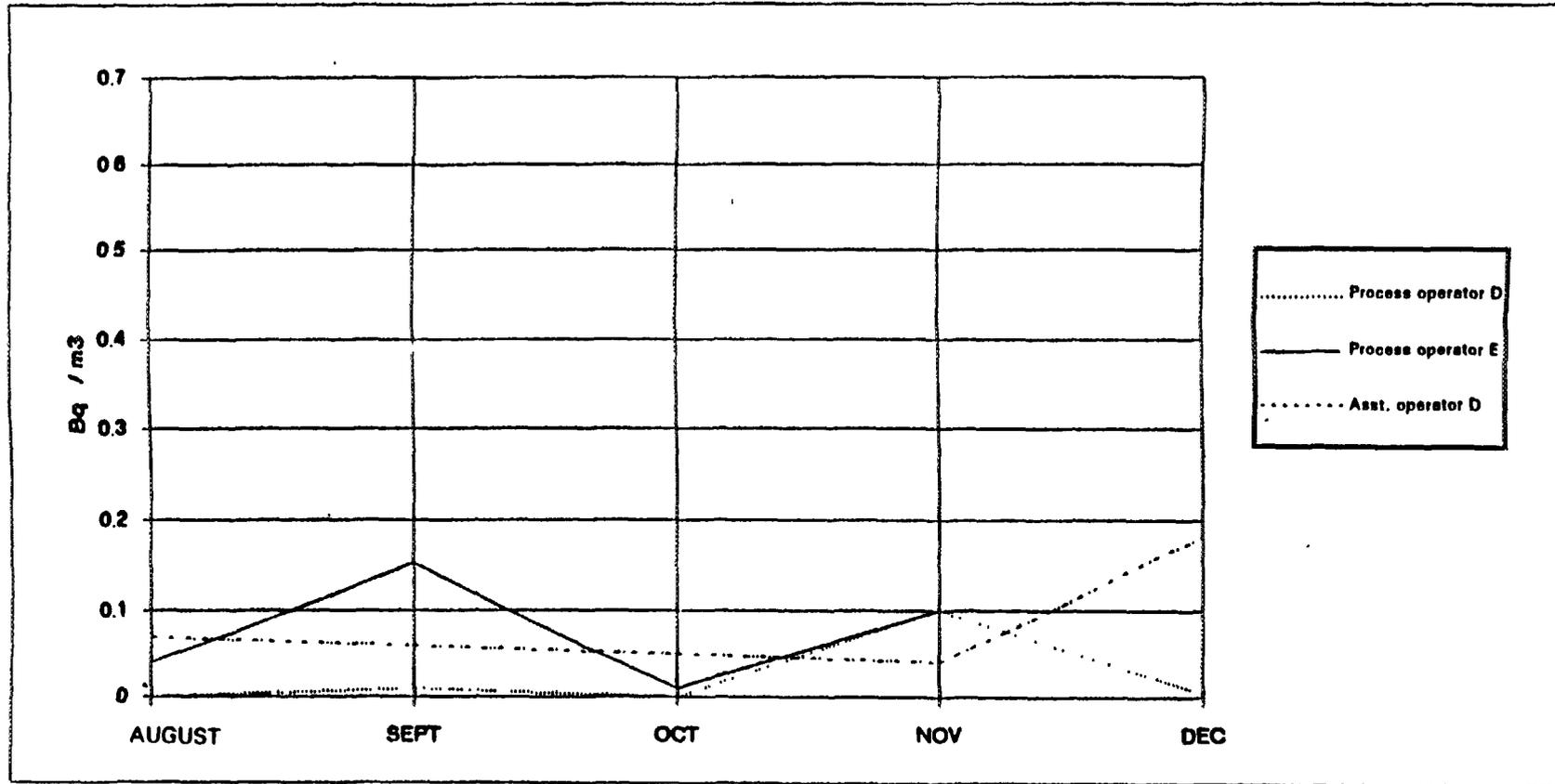
	JANUARY	FEBRUARY	MARCH	APRIL	MAY	JUNE	JULY	AUGUST	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
Drum emptying	0.06	0.92	0.68	0.23	0.14	0.11	0.12	0.16	0.31	0.1	0.2	0.02
Container emptying	0.06	0.09	0.11	0.07	0.07	0.08	0.09	0.11	0.25	0.04	0.07	0.02
First floor	0.05	0.06	0.06	0.03	0.08	0.12	0.18	0.09	0.06	0.16	0.04	0.01
Specific LDCA value	1.12	1.59	1.46	1.54	2.25	5.57	2.24	1.5	1.93	1.13		

FIG. 2a. Digestion Plant - Atmospheric monitoring by aerosol samplers
Monthly averages for 1992



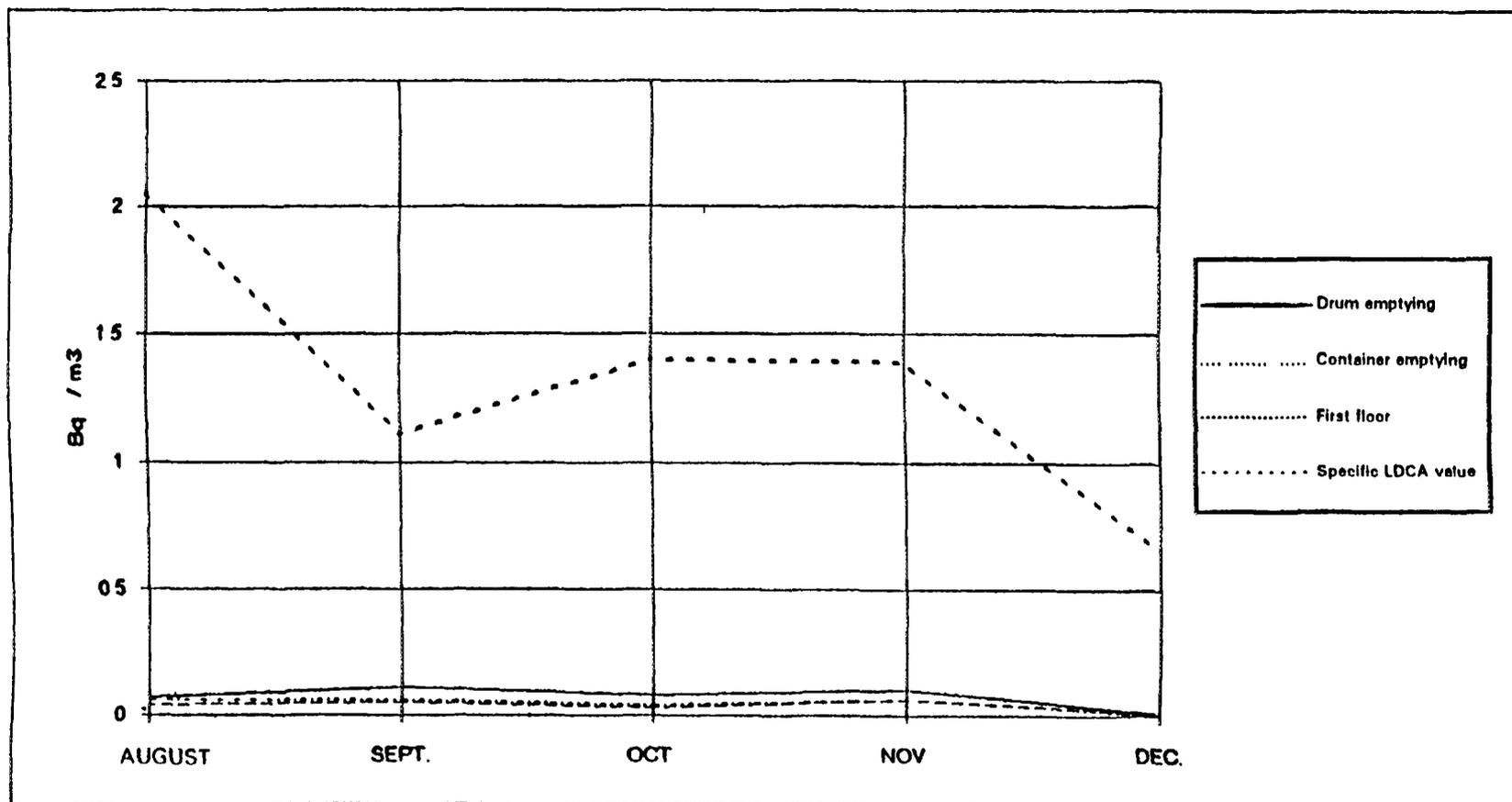
	JANUARY	FEBRUARY	MARCH	APRIL	MAY	JUNE	JULY	AUGUST	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
Drum emptying	0.07	0.09	0.21	0.08	0.09	0.06	0.05	0.46	0.4	0.25	0.11	0.02
Container emptying	0.04	0.04	0.06	0.05	0.06	0.04	0.04	0.11	0.11	0.1	0.05	0.03
First floor	0.07	0.04	0.06	0.08	0.04	0.07	0.04	0.05	0.05	0.08	0.05	0.02
Specific LDCA value	1.58	1.8	2.29	1.98	1.43	3.5		2.72	1.05	1.72	1.42	

FIG. 2b. Digestion Plant - Atmospheric monitoring by aerosol samplers
Monthly averages for 1991



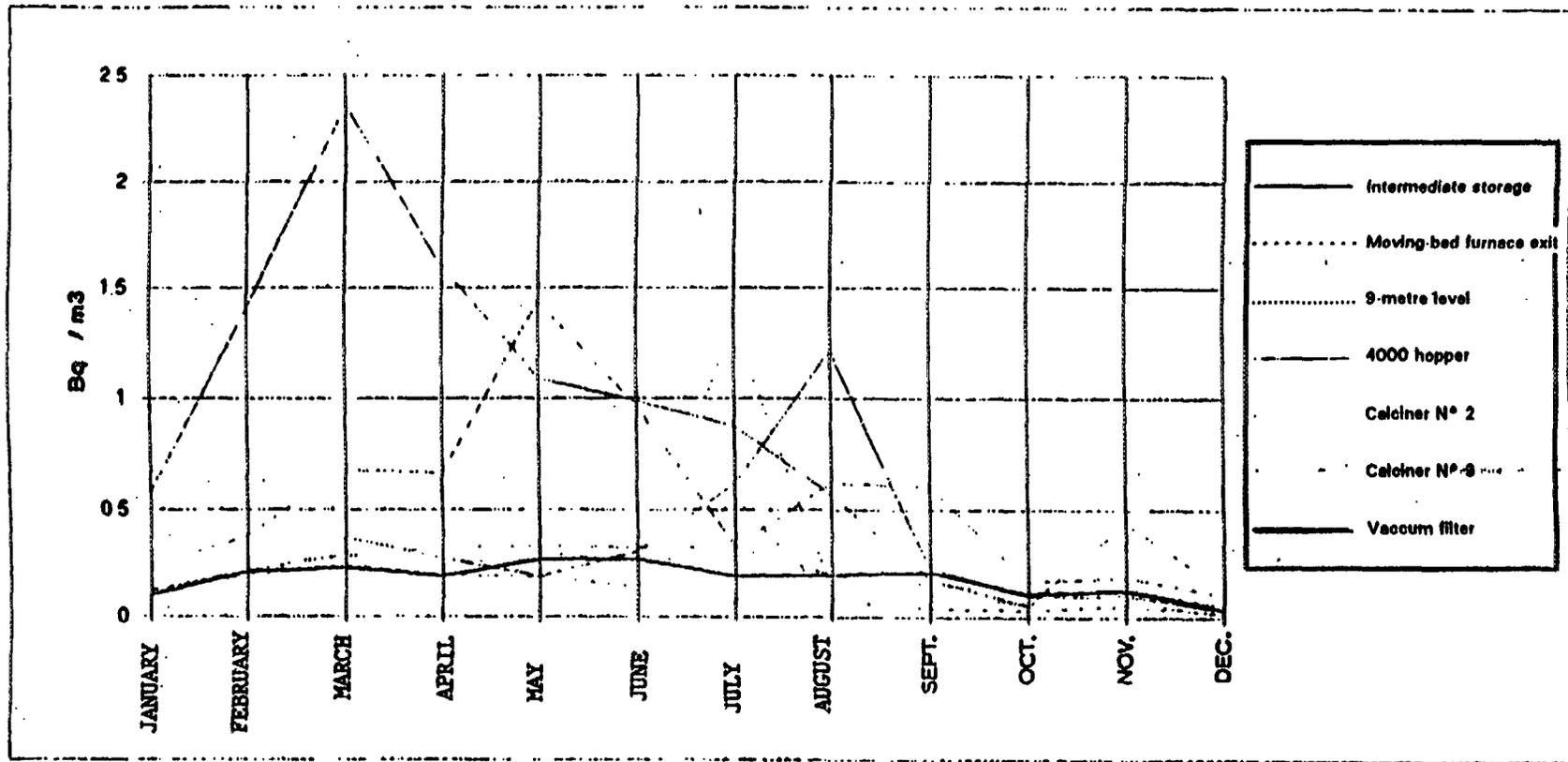
	AUGUST	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
Process operator D	0.00	0.01	0.00	0.10	0.005
Process operator E	0.04	0.15	0.01	0.10	0.18
Asst. operator D	0.07	0.06	0.05	0.04	0.18

FIG. 3. SID1 data for 1990



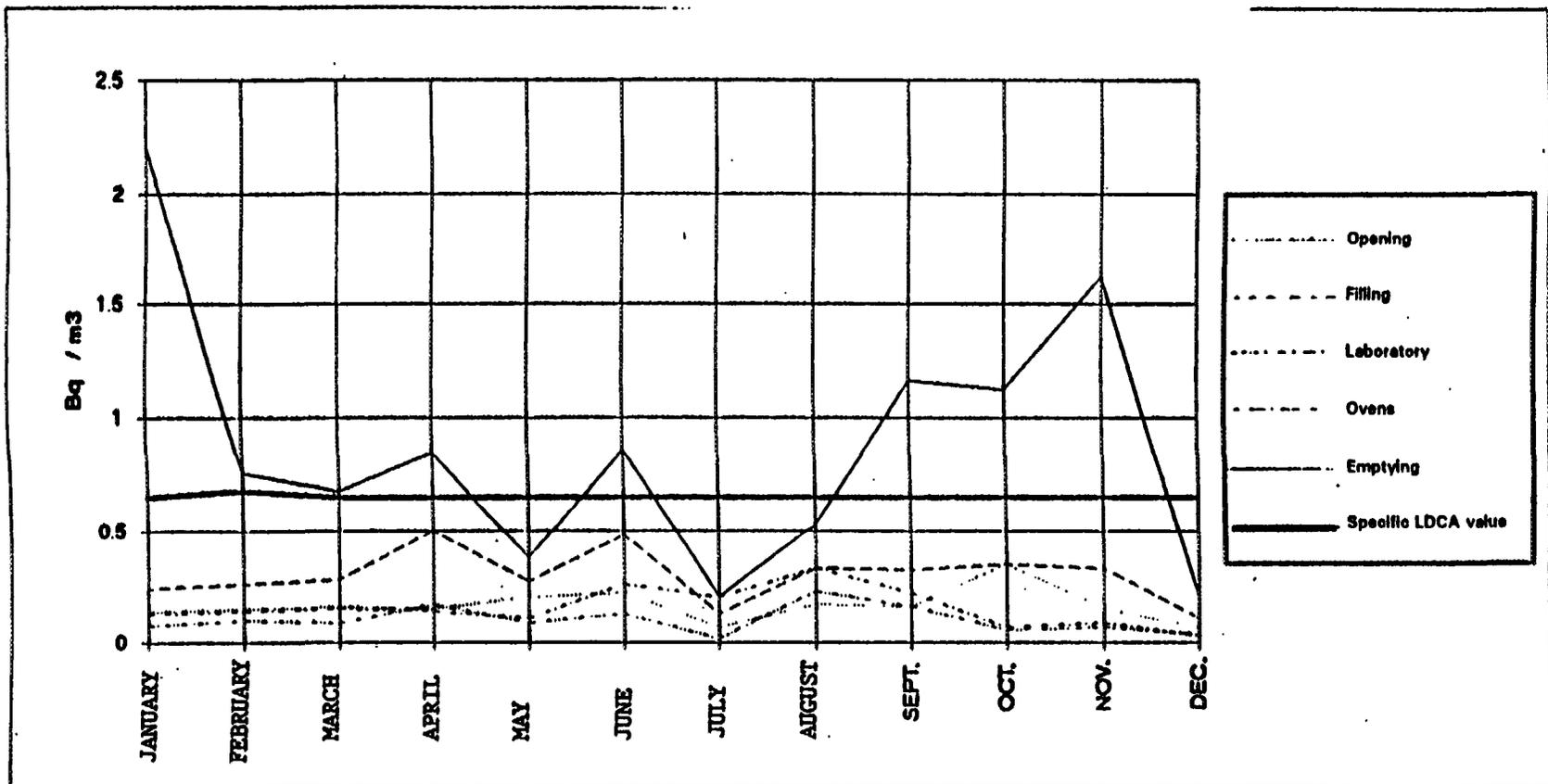
	AUGUST	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
Drum emptying	0.07	0.11	0.08	0.10	0.007
Container emptying	0.06	0.06	0.04	0.06	0.003
First floor	0.04	0.05	0.03	0.06	0.005
Specific LDCA value	2.05	1.11	1.40	1.39	0.65

FIG. 4. Digestion Plant - Atmospheric monitoring by aerosol samplers
Monthly averages for 1990



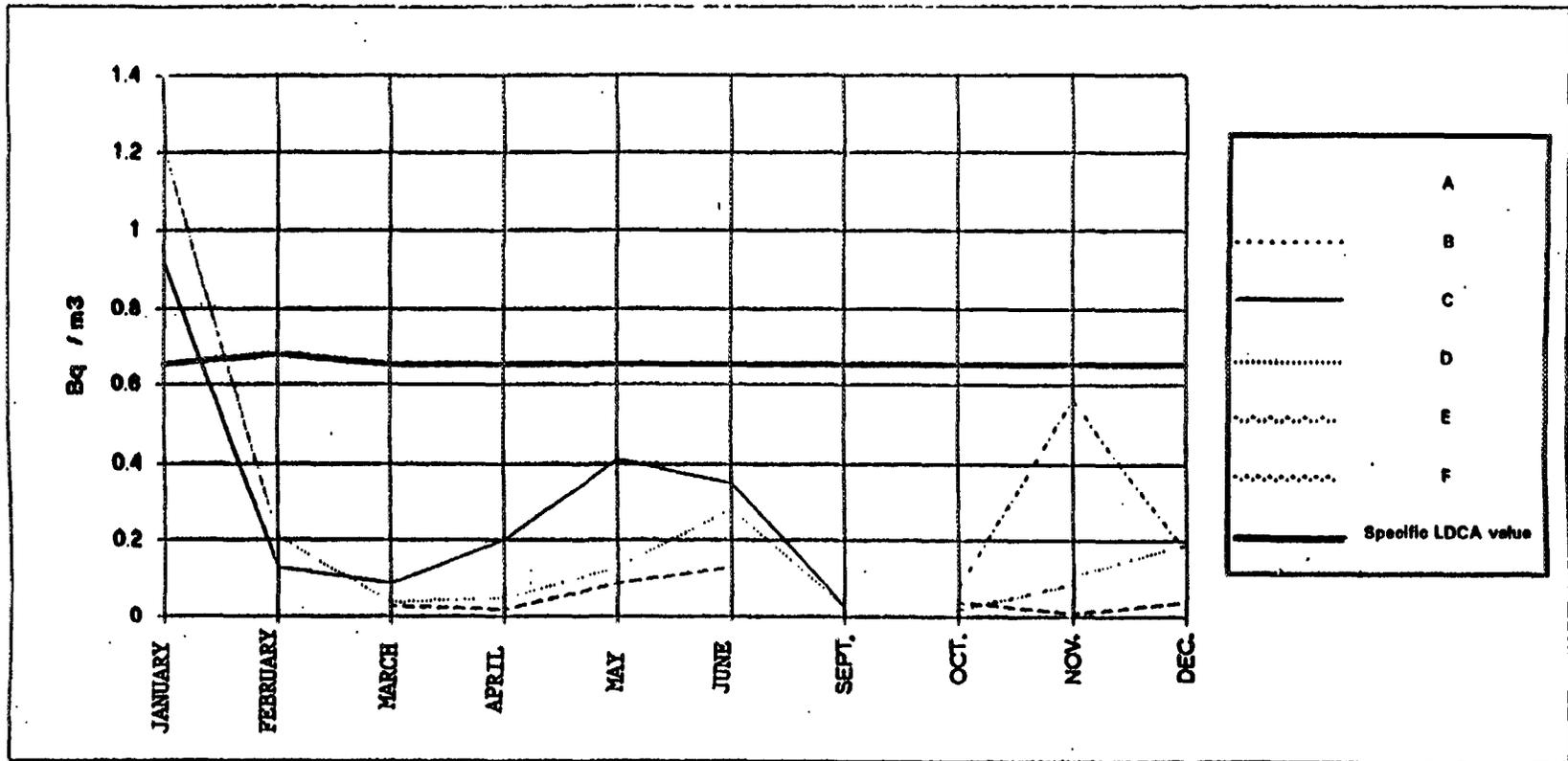
	JANUARY	FEBRUARY	MARCH	APRIL	MAY	JUNE	JULY	AUGUST	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
Intermediate storage	0.58	1.40	2.36	1.59	1.09	0.99	0.88	0.57	0.17	0.05	0.44	0.12
Moving-bed furnace exit	0.12	0.19	0.23	0.18	0.18	0.13	0.55	0.07	0.04	0.03	0.05	0.02
9-metre level	0.20	0.23	0.28	0.23	0.30	0.24	1.32	0.13	0.10	0.06	0.07	0.01
4000 hopper	0.37	0.37	0.37	0.27	0.18	0.30	0.62	1.21	0.22	0.09	0.10	0.03
Calciner N° 2	0.11	0.21	0.28	0.32	0.33	0.32	0.32	0.19	0.15	0.11	0.11	0.05
Calciner N° 3	0.23	0.38	0.68	0.67	1.43	0.97	0.33	0.62	0.61	0.16	0.18	0.10
Vacuum filter	0.09	0.20	0.22	0.18	0.26	0.26	0.18	0.19	0.20	0.10	0.12	0.03

FIG. 5. Precipitation and Hydrofluorination Section
Atmospheric monitoring by aerosol samplers - Monthly averages for 1992



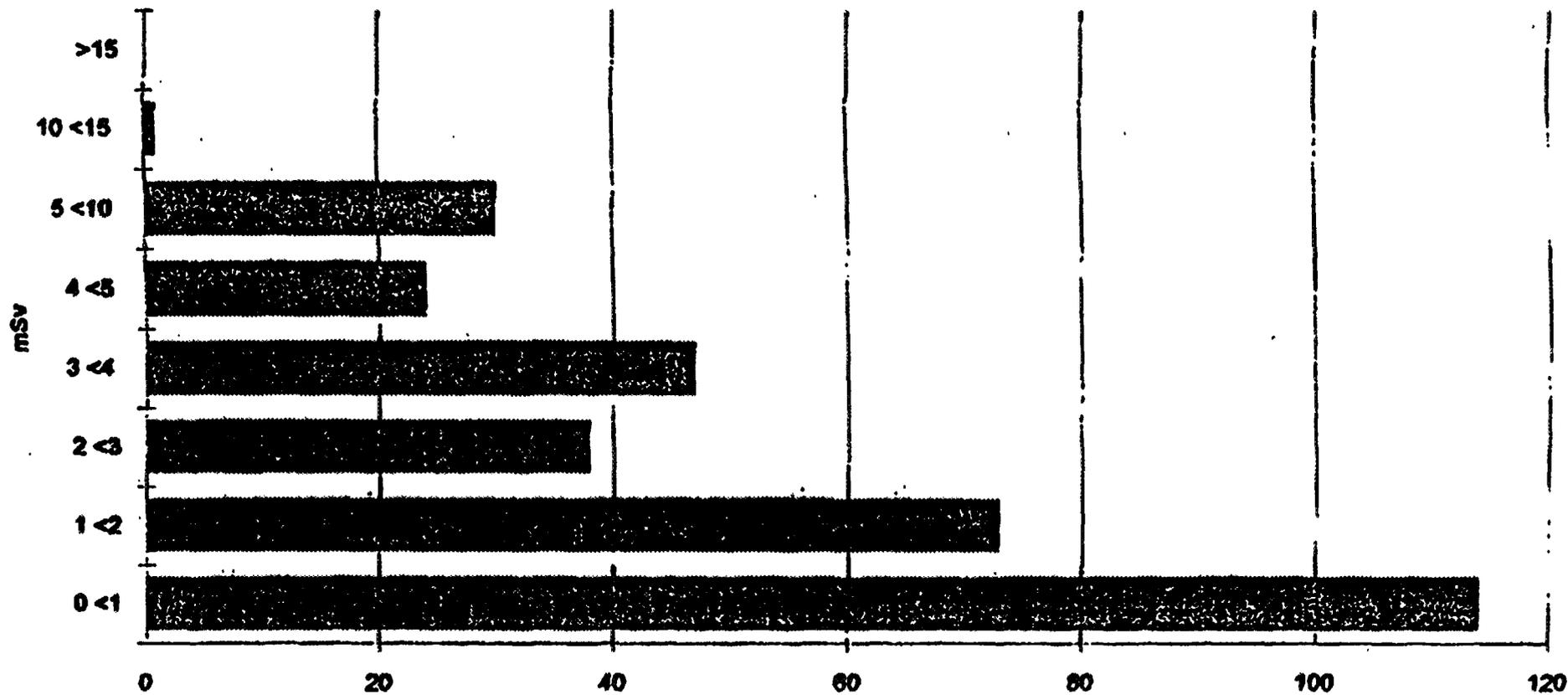
	JANUARY	FEBRUARY	MARCH	APRIL	MAY	JUNE	JULY	AUGUST	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
Opening	0.14	0.15	0.16	0.15	0.20	0.22	0.07	0.17	0.16	0.35	0.15	0.07
Filling	0.24	0.26	0.28	0.50	0.27	0.48	0.13	0.33	0.32	0.35	0.33	0.11
Laboratory	0.13	0.14	0.15	0.14	0.11	0.26	0.20	0.33	0.22	0.07	0.09	0.03
Ovens	0.08	0.10	0.09	0.17	0.09	0.13	0.02	0.23	0.16	0.06	0.07	0.04
Emptying	2.20	0.76	0.68	0.85	0.38	0.86	0.20	0.53	1.16	1.12	1.62	0.20
Specific LDCA value	0.68	0.68	0.65	0.65	0.65	0.65	0.65	0.65	0.65	0.65	0.65	0.68

FIG. 6. *Sampling Station*
Atmospheric monitoring by aerosol samplers - Monthly averages for 1992



	JANUARY	FEBRUARY	MARCH	APRIL	MAY	JUNE	SEPTEMBER	OCTOBER	NOVEMBER	DECEMBER
A	0.		0.27							0.03
B	0.83		0.03	0.02	0.05	0.13		0.04	0.01	0.04
C	0.92	0.13	0.09	0.20	0.41	0.35	0.03			
D	1.21	0.21	0.04	0.06	0.13	0.28	0.03		0.11	0.19
E								0.02	0.09	
F								0.08	0.56	0.17
Specific LDCA value	0.65	0.68	0.65	0.65	0.65	0.65	0.65	0.65	0.65	0.65

FIG. 7. Sampling Station - SIDI data 1992



1993 total : 0.70 man-Sv

FIG. 8. *Comurhex - Malvésí Works*
Potential individual exposures - 1993 : 327 people monitored

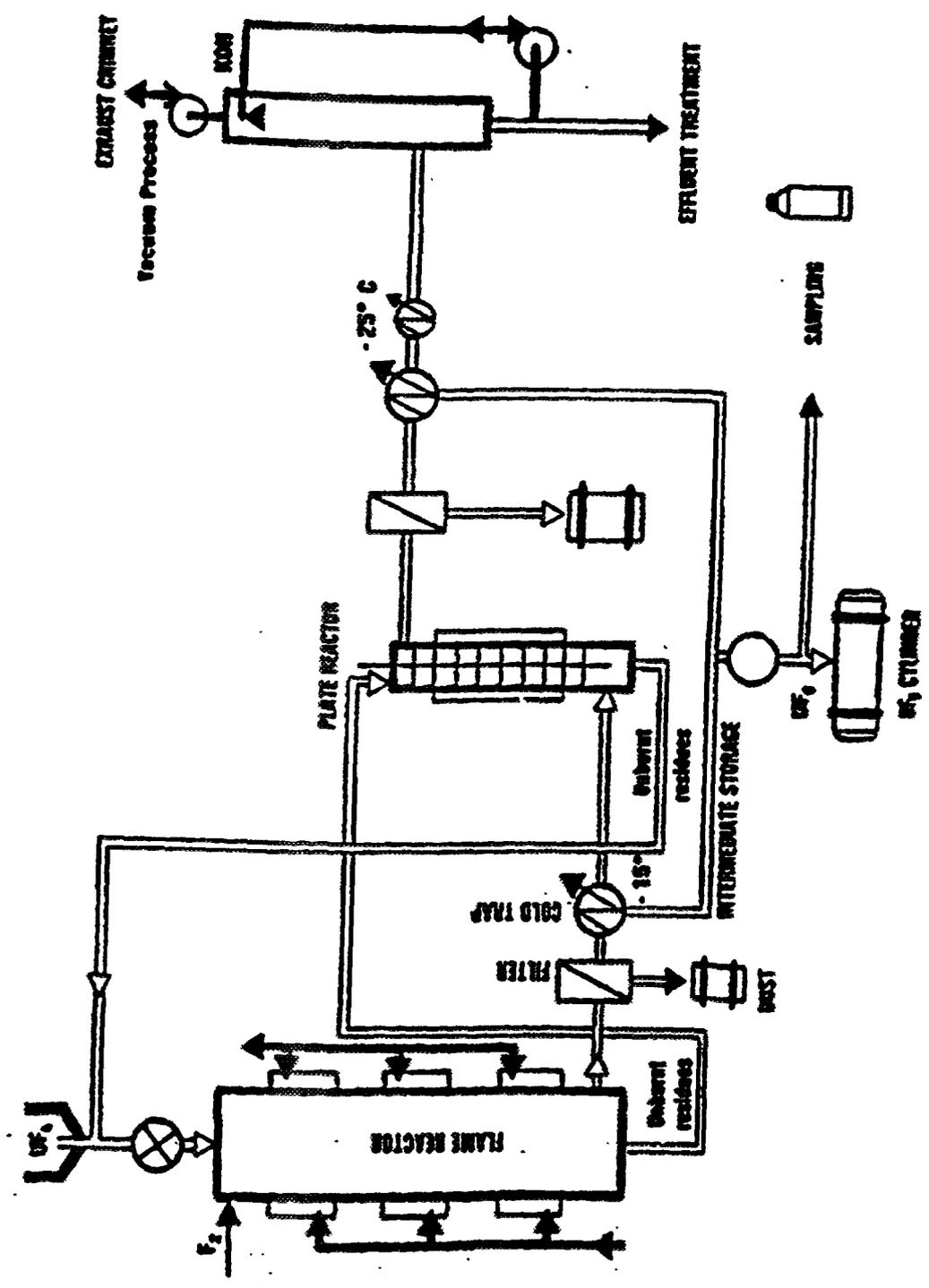


FIG. 9. UF₄ Fluorination

PIERRELATTE

The process of fluorination of natural uranium tetrafluoride (UF_4) carried on in the fluorination plant is detailed in Figure 9.

The tetrafluoride brought up by road-tanker from Malvési to Pierrelatte is conducted by pneumatic conveyor to storage bins.

A pneumatic conveyance system is also employed to feed the flame reactors in which the UF_4 is fluorinated. The reaction is highly exothermic, requiring the reactors to be cooled, and the reaction product, uranium hexafluoride (UF_6), is collected in traps cooled to $-15^\circ C$. Unreacted material and reaction residues are recycled to the reactor inlets, while excess fluorine is burned off in a plate reactor fed with UF_4 and unreacted material.

The entire process is operated under vacuum so as not to endanger the environment, any leakage automatically resulting in ingress of air which is readily detectable by pressure monitoring in association with gas-chromatographic analysis.

Only the liquid UF_6 line from cold traps to containers is pressurised, environmental protection being here assured by the full and complete confinement of this circuit.

The system of supervision operated in the various parts of the Works will be described in due course.

SURVEILLANCE OF PEOPLE AT WORK

Next, to list the means employed in surveillance, both of the worker and of the workstation.

Now, we are concerned here only with radiological protection monitoring and not with other physical, or chemical, hazards to which personnel may be exposed, as exemplified, for instance, by hydrogen fluoride or ammonia, two chemicals employed in quantity at Malvési.

Radiological protection is based on the health physics principles enunciated by the International Commission on Radiological Protection (ICRP), i.e.

- the need to justify the use of ionising radiation when weighed against its potentially damaging radiological implications;
- the need to optimise means of radiological protection, based on the ALARA concept, to ensure that doses or probabilities of exposure are as low as possible for a given set of technical and economic imperatives;
- the need to limit individual doses and risks.

However, as we shall see in a moment, the ICRP 60 guidelines broaden the field as previously established and will be requiring us to make certain changes.

Two aspects have to be considered - workstations and workers - and the means employed are several.

A. Surveillance of workstations and of working environments

Surveillance of workstations and working environments means monitoring potential external exposure and potential internal exposure at the workstation considered.

A.1 *Malvési Works*

Surveillance of exposure to ambient external radiation is warranted only at very few points in Malvési Works, given that natural uranium is only a very weak gamma-emitter, and is exercised by means of three fixed γ -alarm monitors positioned at key locations.

Potential internal exposure by inhalation of uranium dust is monitored by means of aerosol samplers which continuously monitor atmospheric contamination in the various plants and shops making up the Works.

Twenty-five aerosol samplers strategically positioned throughout the Works, at points of maximum exposure, each draw-in air at the rate of 1.25 cubic metres per hour, an intake corresponding to the typical respiratory pattern of a man at work. They are fitted with 140 mm diameter filters which are checked daily for alpha- and beta-emission.

The results of these daily checks are compared with limits of concentration in air (LDCA¹ values) calculated from annual incorporation limits (AILs). The LDCA value is defined as the concentration (expressed as Bq/m³) of a radioelement in air which, if inhaled over the standard total of 2000 working hours, would result in an internal exposure equivalent to the AIL.

A.2 *Pierrelatte Works*

External exposure monitoring is warranted by the concentration, at specific points in the Works, of natural radionuclides resulting from mechanisms of selective fluorination and is carried out by means of fixed environmental monitors, in combination with weekly measurements of irradiation at points of movement of personnel.

Potential internal exposure by inhalation of uranium dust is monitored via a total of 37 aerosol samplers strategically positioned at points of maximum exposure throughout the Works and continuously monitoring atmospheric contamination in the various plants and workshops. Drawing-in air at the rate of 1.25 m³ per hour, corresponding to the typical respiratory pattern of a man at work, these samplers are fitted with 140 mm diameter filters which are checked daily for α and β emission. The results of these daily checks are compared with limits of concentration in air (LDCA values) calculated from annual incorporation limits (AILs) (Figure 10).

The LDCA value is defined as the concentration (expressed as Bq/m³) in air of a radioelement which, if obtaining over the standard total of 2000 manhours, would result in an internal exposure equivalent to the AIL.

¹ Limites Dérivées de Concentration dans l'Air (calculated limiting values of atmospheric concentration).

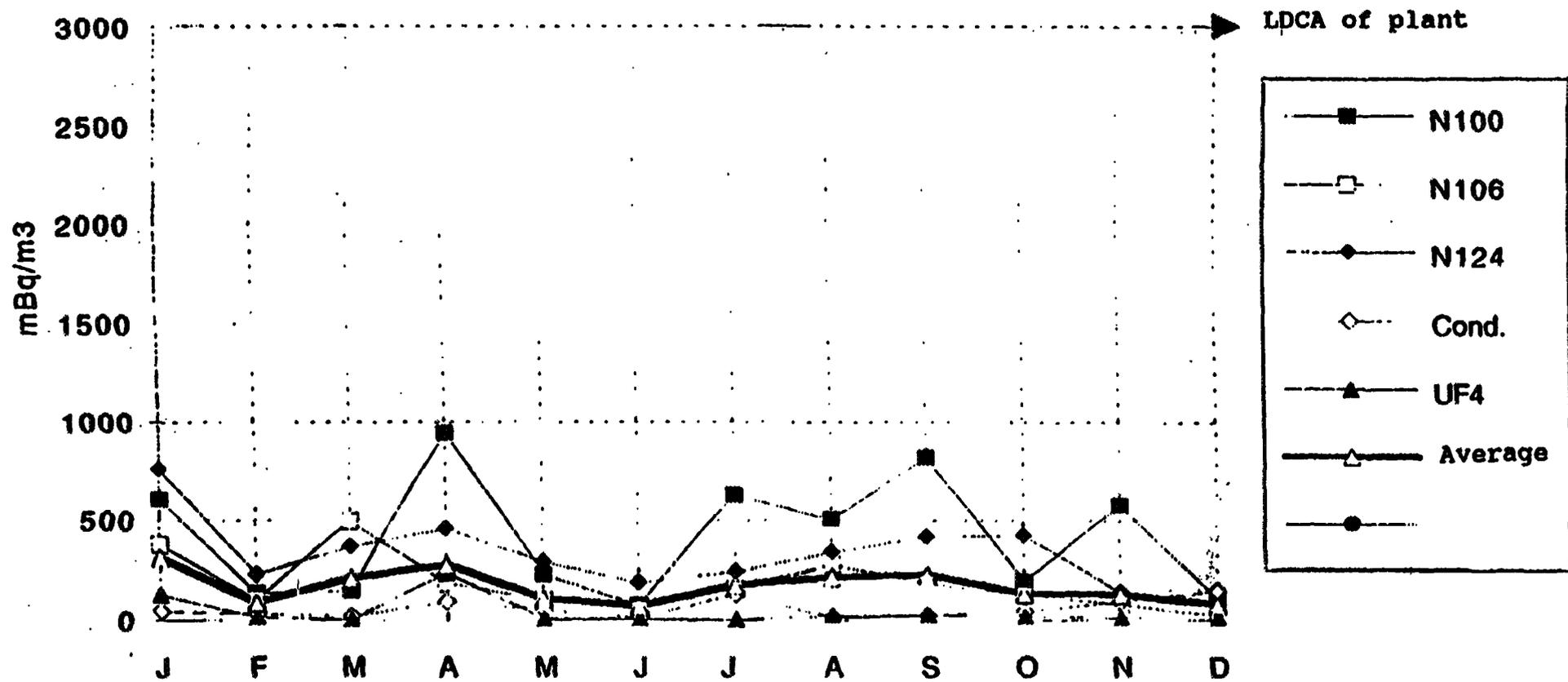


FIG. 10. Fluorination Plant
 Atmospheric monitoring by aerosol samplers - Monthly averages for 1993

“Edgar” alarms are also employed for instantaneous atmospheric contamination monitoring, enabling the appropriate means of individual-operator radiation protection to be triggered if and when required.

B. Surveillance of the workforce

B.1 Malvési

The system comprises surveillance both of external exposure and of internal exposure.

External exposure is measured by means of individual monthly or three-monthly film badges, depending on personnel classification (A or B).

Internal exposure is monitored via determinations of uranium in urine and whole-body γ -measurements.

The workforce is monitored both for external and for internal exposures.

External exposure is measured by means of individual monthly film badges. Internal exposure is evaluated via (i) determinations of uranium in urine, (ii) whole-body gamma measurements and (iii) determinations of uranium in stools.

C. SIDI² system

Another, more particular, means of surveillance employed by Comurhex for certain members of the workforce is the SIDI system. This uses an individual air sampler associated with a measuring head; the monthly-based measurements comprise alpha counts on “long-life” radioactive aerosols and the detection of “short-life” alpha-emitter aerosols (incorporating radon decay products).

SETTING PRACTICAL TLV VALUES

Now to discuss how the overall system of surveillance is organized and how the TLVs adopted at Malvési have been determined.

Genuine and thorough familiarity with the processes employed, with process technologies and operating conditions, and with the precise activity engaged in (ergonomically speaking), is absolutely indispensable.

This is because we are dealing, not with conditions in the laboratory, but with industrial processes, i.e. where departures from ideal conditions and a variety of incidents can occur, with potential implications for process operation and, hence, the quality of the end-product: its chemical nature, particle size and content, if any, of impurities.

² Système intégré de dosimétrie individuelle (integrated individual dose measuring system)

Also to be borne in mind is that any toxic chemicals present (e.g. gaseous ammonia or solvents) could compound the toxicity of uranium by modifying the body's natural clearance functions (renal and bronchial).

The three major parameters strongly influencing levels of retention/excretion of uranium are:

- i) transferability (or solubility), as determined by the chemical nature of the substance concerned;
- ii) particle size, as measured by median aerodynamic diameter (DAMA³);
- iii) modes of contamination, i.e. acute or chronic.

The TLVs set in practice will, therefore, vary with conditions peculiar to particular parts of the Works.

Surveillance can be a relatively simple matter where all the people concerned always work in the same shop or plant and at identical workstations. Complications set in when they move around the Works (as in the case of supervisors, electricians, fitters, inspectors and the like).

There can therefore be no single, all-embracing, approach to surveillance, but only approaches specific to particular parts of the Works or, in some cases, to the person.

For each phase of the conversion process, therefore, the approach to surveillance and TLVs is determined on the basis of, and by reference to:

- complete familiarity with the process and process technology;
- the mode of contamination, acute or chronic, normal operating conditions being associated with low-level chronic contamination;
- theoretical knowledge of how the substance considered behaves in the organism (ICRP data);
(Note: we are concerned here solely with natural uranium.)
- practical knowledge based on studies of workstations and physicochemical and biological data - the chemical nature of the uranium, concentration, impurities, hydration, particle size and solubility as determined by tests yielding reliable and readily reproducible results.
(E.g.: technical uranium tetrafluoride is only some 95 - 96% pure and will contain impurities such as uranium dioxide and uranyl fluoride, UO₂F₂.)

Work has already been done in this connection at Malvési and the results of these studies are now of vital importance to health physics applied to the industrial environment.

ICRP 60 will mean applying an annual-exposure limit averaging 20 mSv over a period of five consecutive years, while not exceeding 50 mSv in any one year at Malvési.

³ Diamètre aérodynamique médian en activité (median aerodynamic diameter in activity)

In most of its installations, Comurhex are already operating at below this limit, which can however sometimes be approached if doses are estimated from the theoretical ICRP data (solubility /1 μ particle size), whereas familiarity with the workstations and precise determination of the physicochemical characteristics of the substance or substances concerned yield lower estimates.

~~Let us take a typical case~~, on the basis of which to discuss, for the plant concerned, the system of surveillance operated and TLVs adopted, and then go on to consider particular features encountered in other sections of the Works.

1. The Digestion plant

The personnel working in it are specific to this section, which takes in concentrates of two kinds:

- oxides, featuring a DLCA value, for 1 μ particle size, of 0.65 Bq α/m^3 ;
- uranates, featuring a 1 μ DLCA of 20 Bq α/m^3 .

Both the above types of concentrates will be being processed at any one time.

1.1 Chronic contamination scenario

AIL values specific to the plant are calculated once a month (by the officially prescribed procedure), based on uranate and uranium dioxide throughputs for the plant.

However, since these AIL values are, by definition, known only *a posteriori*, they obviously cannot be used to determine the alarm thresholds.

The thresholds for the Digestion plant have been established on the basis of the LDCA value for the oxides (0.65 Bq α/m^3 rather than for uranates.

The criteria adopted as indicating the need for remedial action are:

- a) Any significant deviation from the normal operation of the plant, which has a total of four aerosol samplers variously positioned:

at the drum-emptying station (working by suction), for which the TLV is 0.4 Bq α/m^3 ;
at the container-emptying station,
at the sampling station (since January 1993) and
on the first floor, at the filters, for each of which the TLV is 0.2 Bq α/m^3 .

- b) Any steady drift in sampling results, even though the relevant alarm threshold may not actually have been reached.

Monthly-average aerosol-sampling data for 1990, 1991 and 1992 are charted in Figure 2.

The medical alarm threshold has been set at 0.65 Bq α/m^3 , i.e. at the LDCA for the oxides.

The SIDIs worn by certain members of personnel enable it to be checked that aerosol samplers have been correctly positioned in the plant, i.e. as near as possible to potential points of emission and therefore recording higher results than SIDIs (cf. Figures 3 and 4).

All personnel working on the Digestion plant are monitored via:

- their external exposure badge;
- regular determinations of uranium passed in urine;
- whole-body gamma measurements.

If the precise physicochemical characteristics of the uranium were known, the AIL value specific to the plant could be arrived at.

1.2 Potential acute contamination scenario:

Typical causes of acute contamination could be incidents such as spillage from an upset drum or leakage when uranium is being sucked from a drum or container.

The material could then be scattered around the Digestion section, but would consist only of a single type of concentrate, oxide or uranate, for which the theoretical LWA value is known, i.e.:

- 0.65 Bq α /m³ for oxides;
- 20 Bq α /m³ for uranates.

Personnel would then apply the prescribed remedial procedure (including the use of special equipment) and be placed under medical supervision (initially involving determinations of uranium in urine).

OTHER SECTIONS OF THE WORKS

2. Purification

On this plant, operating the liquid-phase purification process, there is no exposure to dust. There could, however, be a risk of external exposure when working on raffinates, where deposits trapping thorium and protoactinium-234 collect.

Under normal operating conditions, the Purification section is no more than a walkthrough zone, containing no workstations as such.

A γ -radiation detector is located in proximity to the each raffinate and set to trip at 25 μ Gy/h, which corresponds to 50 mSv for a total of 2000 working hours.

If the measured value exceeds 5 mGy/h, entry into the zone is prohibited. If it lies between 25 μ Gy and 5 mGy/h, certain precautions have to be taken when carrying out remedial action (cleaning) and working time in the zone during the month is restricted so as not to exceed 2.5 mSv/month. This is an in-house rule at Malvésí and equates to a maximum level of exposure of 2.5 x 11 (months) = 27.5 mSv/annum.

Note that the collective dose figure is unaffected by these calculations, which refer solely to a walk-through zone and not to a workstation.

3. Precipitation and Hydrofluorination

The people working on the Precipitation and Hydrofluorination plants are exposed to uranium trioxide and tetrafluoride dusts, two compounds theoretically classified "W".

Now, physicochemical investigation has established that their behavioral classification should in fact be closer to "D". The LDCA value is therefore 10 Bq α/m^3 in radiological terms, but the TLV applicable here has to do with chemical toxicity and is therefore lower, at 6.3 Bq α/m^3 .

Technically-determined TLVs

There are seven monitoring points, variously employing:

3 aerosol samplers, with TLV set at 0.8 Bq α/m^3

Precipitation: at the Calciner No. 2 exit
at the Calciner No. 3 exit

Hydrofluorination: at the point of intermediate storage.

4 aerosol samplers, with TLV set at 0.4 Bq α/m^3

Precipitation: at vacuum filter No. 2

Hydrofluorination: at the moving-bed furnace exit
at the 4000 hopper
at the 9 metre level.

(cf. Figure 5)

Medically determined TLVs

2.1 Bq α/m^3 (i.e. one-third of 6.3 Bq α/m^3).

Personnel working on these two plants are subject to surveillance via:

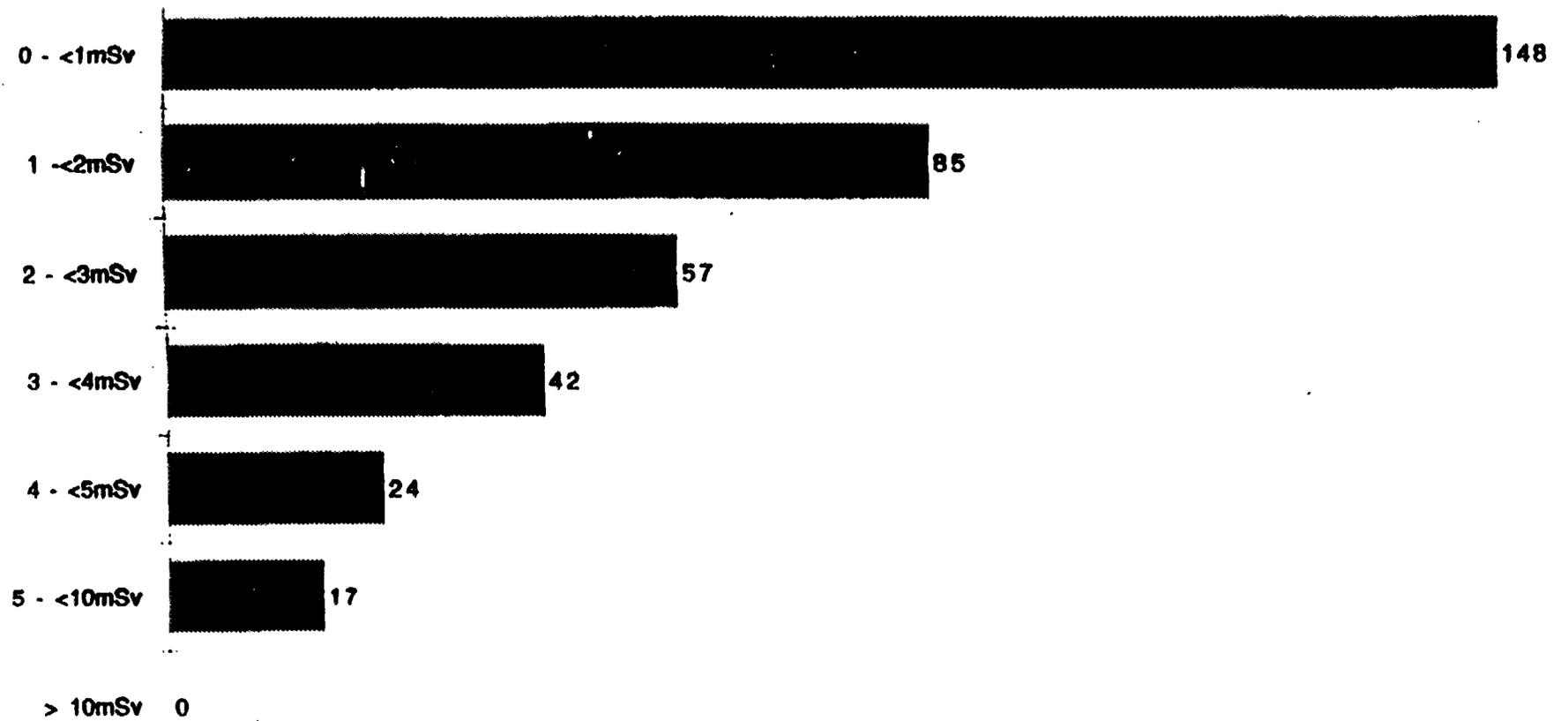
- external exposure badge;
- determinations of uranium excretion in urine.

4. Recovery

The approach applied on the Recovery plant is exactly as just described.

5. Sampling station

The Sampling station constitutes a special case by virtue of the particular features of the operating procedures employed (drum inversion and dust extraction at low vacuum to avoid modifying the characteristics of the material being sampled).



*FIG. 11. Comurhex -Pierrelatte Works
Potential individual exposures - 1993 : 373 people monitored*

The materials sampled are uranium oxide and uranates and the alarm threshold is 0.65 Bq α/m^3 (the LDCA for oxides).

The drum-inversion installation in the Sampling station generates dust at a rate oscillating about this alarm threshold and is so positioned that it contributes significantly to airborne dust in the shop as a whole.

It is planned to introduce a means of confinement of the drum-inversion installation in response to the CIPR's request that exposure be kept to the minimum reasonably achievable. This with a view to the new CIPR 60 standards of protection (20 mSv per annum over a 5- year period). (Cf. Figures 6 and 7.)

AT PIERRELATTE

The position in our Pierrelatte facilities is that, for all our personnel, we are well below half the proposed ICRP 60 limit.

Personnel performing more than one function move around the entire uranium sector with no compartmentation of process and other units.

Zones are defined according to the level of classification (monitored or controlled). Due account has to be taken of the LDCA of uranium tetrafluoride and, in order to allow for the presence of Category-B personnel, we have lowered the theoretical value from 10 to 3 Bq α/m^3 .

The Edgar alarms monitoring instantaneous levels of atmospheric contamination alert personnel to the need to protect the respiratory tract (by donning masks).

The strategically positioned aerosol samplers enable the internal doses received by individuals to be evaluated (evaluations erring on the high side in taking no account of respiratory protection) (Figure 10).

Whenever any operation involving the opening-up of process plant or equipment is contemplated, a set of strict technical and medical procedures has to be observed:

- respiratory protection is obligatory and a radioprotection specialist or specialists must be present;
- radiotoxicological examinations of urine are mandatory.

ANNUAL RADIOLOGICAL PROTECTION PERFORMANCE

The bar-charts in Figures 8 and 11 record potential individual levels of exposure for Comurhex and contractors' personnel. Potential individual exposure is determined by summing external exposure and internal exposure evaluated from aerosol sampling data.

During the year 1992, 90% of personnel received less than 5 mSv per annum.

With a view to improved monitoring of installations and of the workforce and the future application of ICRP 60, a number of projects are currently being implemented:

- a real-time alarm monitor and preset TLV for major maintenance operations;
- computerization to improve the processing of aerosol sampling data;
- the use of a software package (LUDEP) affording good estimates of internal doses, and work on correlating estimates based on aerosol sampling data with the results of determinations of uranium in urine.

CONCLUSION

Aerosol samplers and Edgar alarms have a vital role to play in the monitoring of the various sections of the Works and, therefore, of personnel.

However, certain categories of personnel - supervisory personnel, electricians, fitters, inspectors and the like - are not fully covered by this mode of surveillance because they have to move about in the Works and because the nature of their duties can sometimes mean that they are more exposed than others. In their case, at Malvési, the SIDI system makes realistic assessments of exposures possible; all the same, the only means of determining their true level of exposure is by determinations of uranium in urine and whole-body gamma measurements.

Our concern has to be to bring the sum total of knowledge, resources and facilities to bear to minimise personnel exposure and also minimise margins of uncertainty affecting the evaluation of internal doses.

This can only be accomplished if we are aware of the precise physicochemical characteristics of the materials being dealt with.



ENVIRONMENTAL SAFETY OF REPROCESSING PLANT LA HAGUE

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Abstract

The La Hague plant (UP2.800 and UP3) was designed and constructed to reprocess spent fuel. This report gives an evaluation of the health and environmental aspects of a reprocessing plant.

1. Introduction

The choice of a specific fuel cycle depends on several different factors: economics, security of supply and also environmental impacts.

The La Hague plant (UP2.800 and UP3) was designed and constructed to reprocess spent fuel; its current capacity is of 1600 tons per year by August 1994. One of the principal concerns of nuclear operators is to ensure that radiation protection standards are complied with, and especially to control environmental-related problems.

2. Basic process [2]

The first La Hague plant (UP2) started up in 1966, a new plant (UP3) started up in 1989 and the latest, a new UP2 plant (UP2.800) started up in 1994. The design of the UP3 and UP2.800 plants is based on the well-known Purex process. After dissolution in nitric acid, plutonium and uranium are separated and thoroughly purified by solvent extraction cycles, using the selective extraction properties of TBP (tributyl phosphate). The fission products are left in the primary nitric solution, which is then concentrated. The resulting stream is calcinated and solidified in a borosilicate glass matrix by the vitrification process.

However, significant process innovations have been specially developed and integrated into the plant with other new technologies. These include the following processes and technologies:

- The process flow-sheet includes total recycling of nitric acid and TBP solvent regeneration by distillation.
- All waste streams are processed and conditioned on line, leading to end products of controlled quality (ie glass, bitumen and concrete for, respectively, fission products, precipitated effluents, and fuel assembly hulls).
- Glass matrix in the new vitrification facilities can incorporate all kinds of high level waste separated by the process: alkaline as well as acid concentrated solutions and undissolved particles from the dissolution step.
- The design of segmented hot cells, standardized equipment (eg pumps, valves, filters etc) and mobile replacement casks ensures efficient remote handling, simplified maintenance procedures and very low radiological exposure.
- New technologies include the horizontal shearing of fuel assemblies, the continuous zirconium dissolver and pulsed annular columns on the high level extraction cycle.
- The whole plant is controlled from one central room where all mechanical and chemical operation are monitored and controlled through a Total Data Management System.

3. Regulatory Requirements [3]

Before the start up of a commercial nuclear facility, the impacts of facility operations on the surrounding environment and, more specifically, on the public must be carefully assessed. This assessment addresses the health effects of plant operations under both normal and accident conditions, and serves as the basis for official French regulation, which establish release limits to be respected at all times. The regulations concern both liquid and gaseous releases, each of which have four activity thresholds for specific elements:

liquid releases: tritium, beta emitters (excluding tritium), strontium - 90 and caesium - 137, and alpha emitters;

gaseous releases: gases other than tritium, tritium, halogens, and aerosols.

Annual release authorization			
Tritium	37.000 TBq	Gases (other than tritium)	480.000 TBq
Beta emitters (excluding tritium)	1700 TBq	Tritium	2200 TBq
90 Sr and 137 Cs	220 TBq	Halogens	110 GBq
Alpha emitters	1,7 TBq	Aerosols	74 GBq

The Office pour la Protection contre les Rayonnements Ionisants (OPRI), or Radiation Protection Office, of the Ministry of Health is responsible for verifying compliance with the release limits. Compliance is determined through radiological monitoring of the site and its environment. Results of radiological measurements which trace the evolution of the environment are sent to the OPRI to be checked and analysed.

4. Process and safety performance

a) Solid Wastes [2]

An ambitious programme of waste volume minimisation is currently underway. It applies to each waste stream of the plant: fuel assembly structural materials (end-fittings and hulls) are grouted with cement; precipitated sludges from the liquid effluents are conditioned with bitumen; various wastes from plant operation are conditioned in concrete.

Hulls and end-fittings. Cogema is currently assessing a hull compaction technology to replace the cementation process, in order to reduce the volume of this waste stream. The pellets produced by compaction will be loaded in a canister identical in shape to a glass canister. This new conditioning technique is intended to be implemented before the end of the decade, producing a final volume of 0.15 m³/t instead of 0.6 m³/t.

Waste from operations. This low and medium level waste is all conditioned in concrete, the waste coated with cement that is strengthened with metal fibres; a very large reduction in volume has been achieved by comparison with design estimates. The main factors have been the use of highly reliable equipment, which reduces the number of maintenance operations, and improved sorting at the entrance of each production facility.

Further minimisation will come from new alpha decontamination techniques, based upon the oxidizing dissolution of plutonium.

Bitumen waste. The excellent separation achieved allowed Cogema to implement complementary improvements in waste management which are aimed at completely suppressing the production of bitumen waste drums by the year 1995. These improvements include:

A more sophisticated segregation of the liquid waste, according to their chemical and radioactivity levels.

- The incorporation of additional evaporation capacity in the plant.
- The appropriate treatment of laboratory effluents which contain a mix of chemical reagents that are not present in the plant process streams.

When all these new units are available, it will be possible to concentrate and to route practically all the activity towards the vitrification facility. The need for precipitation and bitumenisation will thus disappear in normal operation in the intermediate and low level liquid waste. The activity concentration is much higher in glass than in bitumen, so the resulting small increment of activity that will be incorporated in the glass will not induce any noticeable volume increase.

The table compares design volume estimates, current achievements and target values for the end of the decade for all types of waste. If all high level and transuranic wastes are added together, the total volume, which is to be placed eventually in future underground repository, appears already smaller than the volume which would result from the direct disposal of spent fuel (ie. 1.5-1.7 m³/t, according to recent estimates). By the end of the decade, it should be reduced to less than 0.5 m³/t.

Quantities of conditioned waste at UP3					
Nature of waste	Conditioning form	Design value (litre/tonne)	Actual result (1993) (litre/tonne)	Target value (1995) (litre/tonne)	Target value (2000) (litre/tonne)
Waste not compatible with current surface disposal					
Fission products	Glass matrix	130	115	115	115
Hulls and end-pieces	Concrete	600	600	600	150
Precipitation sludge	Bitumen	630	450	0	0
Waste from Operations	Metal fibre strengthened concrete container	1700	200	200	200
Total		3060	1365	915	465
Waste suitable for surface disposal					
Waste from operations	Metal fibre strengthened concrete container	3800	1400	1400	1400

b) Liquid discharge [6]

Concerning the monitoring of gravity drainage systems, the plant has four independent systems to collect the specific effluents:

- rain water
- chemical water from the installations
- domestic water from buildings
- rain water which falls on specific zones, for example on the storage casks.

The first three are discharged into streams, whereas the fourth is released through a pipe into the sea. However, all four types of liquid waste are monitored continuously by sensors which measure the beta and gamma activity.

Liquid waste discharge into the sea

After treatment of the active liquid waste by evaporation and then vitrification some low level liquid waste is discharged into the sea. For that there is a special procedure: we can release the waste,

only at high tide, during a period of 3 hours; that is, two and a half hours before the high tide, and half an hour after it. All the effluents are analysed by the laboratory before release. In order to release the waste, it is necessary to get authorization from the director. We can authorize the release, according to the norms in relation to the analyses.

The results for the year 1993 are:

Annual liquid release in 1993			
Radionuclides	Activity (MBq)	% of the authorization	Normalized release Bq/MWe/yr.
Tritium	5,15 10 ⁹	13,9%	2,4 10 ¹¹
Beta emitters (excluding tritium)	7,32 10 ⁷	4,3%	3,4 10 ⁹
90Sr + 137 CS	2,89 10 ⁷	13,1%	1,3 10 ⁹
Alpha emitters	1,01 10 ⁵	5,9%	4,7 10 ⁶

c) Gaseous releases [5]

For gas discharges, the monitoring strategy is composed of three concentric circles centred on the plant.

- the main chimneys, UP2 400, UP2 800 and UP3 are at the centre.
- the parameters to be measured are those required by the gaseous waste authorization decrees:
 - alpha aerosols
 - beta aerosols
 - rare gases (krypton - 85)
 - and the rate of stack discharge.

We also measure tritium and iodine.

- The second circle for monitoring waste gases consists of eight stations placed at the boundaries of the plant, where we control beta aerosols, and gamma radiation.
- The third circle consists of five measuring stations situated in neighbouring villages around the site at distances of 1 to 6 km where we also control:
 - gamma radiation
 - beta activity of gas
 - aerosols, iodine and tritium

Annual Gaseous release in 1993			
Radionuclides	Activity (Bq)	% of the authorization	Normalized release Bq/MWe/yr.
Gases	1,15 10 ¹⁷	23,97%	5,3 10 ¹²
Tritium	4,21 10 ¹³	1,92%	2 10 ⁹
Halogens	1,08 10 ¹⁰	9,86%	5 10 ⁵
Aerosols	8,97 10 ⁶	0,012	4,2 10 ²

5. Potential Health and Environmental Impacts during routine operation

a) Occupational [1]

The average annual dose to operating personnel was 0.42 mSv in 1993 below Cogema's design criteria of 5 mSv/yr and well below the maximum allowable dose of 50 mSv/yr. Specific personnel exposure has been reduced by a factor of 30 in the last 15 years to the current level of 0.10 mSv/GWe/year for the exploitation of the plants.

Occupational exposure for La Hague's Plants in 1993			
Monitored workers	Annual collective dose (man Sv)	Annual effective dose per monitored worker (mSv)	Normalized collective dose for exploitation (man Sv/GWe.yr)
8132	3,42	0,42	0,10

Within this total, the UP3 plant contributed only a few percent largely due to the remotely maintainable facility design and to the extensive use of automation in plant operations.

b) Environmental

Environmental health and environmental impacts can be estimated through radiological monitoring.

Radiological monitoring involves taking representative samples from the environment in a regular and periodic programme, and analysing them in the Environmental Laboratory operated by the Radiation Protection Department. The environmental monitoring programme enables detailed dose calculations to be established; it involves some 17,000 samples a year taken from the three pathways for radionuclide migration to the food or biological chain - atmospheric, hydrogeological, and marine - and around 55,000 analyses.

Atmospheric pathways

Continuous real-time monitoring is supplemented by measurements taken on the filters and activated-carbon traps at each plant outlet and at the monitoring stations on the site boundary and in the outlying areas. Monitoring includes potential fall-out in rainwater, vegetation, crops, milk and meats.

Hydrological pathways

The 32 springs and streams originating near the plant are monitored and analysed. Ground water through a site network of 220 piezometers and drinking water can be closely monitored. All drinking water in the district is also regularly analysed.

Marine pathways

A 6 kilometer long submerged pipeline carries liquid releases from the plant out to the sea. The pipeline is regularly inspected. Two hundred kilometers of coastline, from Granville to Le Havre, are sampled, including water, sand, sediment, crustaceans, shellfish and seaweed which act as filters for radionuclides released into the sea. Deep sea sampling includes water, sediments and fish. Sampling is performed by the French Navy, while the Marine Radioecology Laboratory of the French Atomic Energy Commission (CEA) studies marine dispersion of radionuclides in the English Channel and the North Sea.

[4]

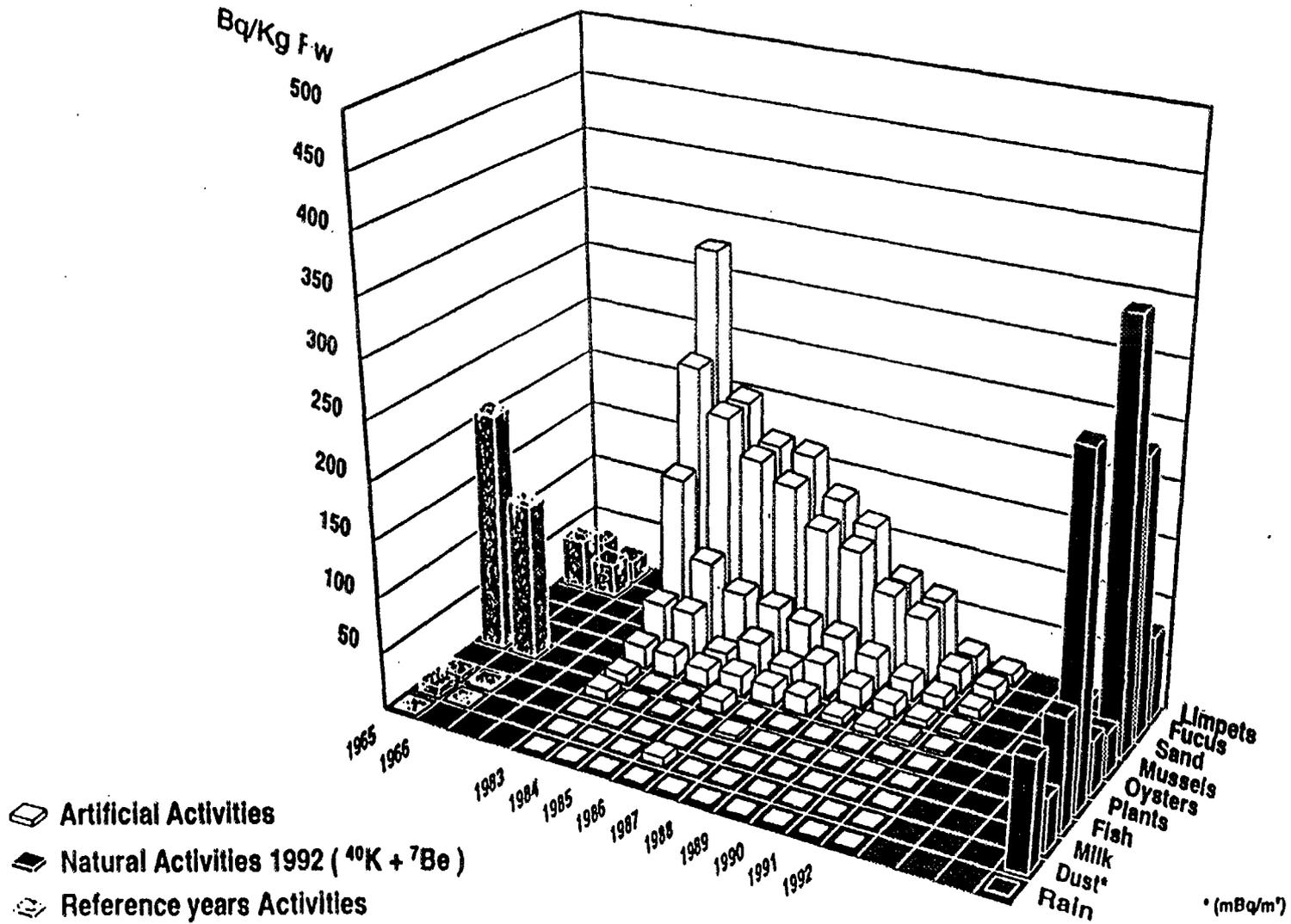


FIG. 1. Environmental monitoring data for the last 10 years together with measurements taken in 1995 & 1996.

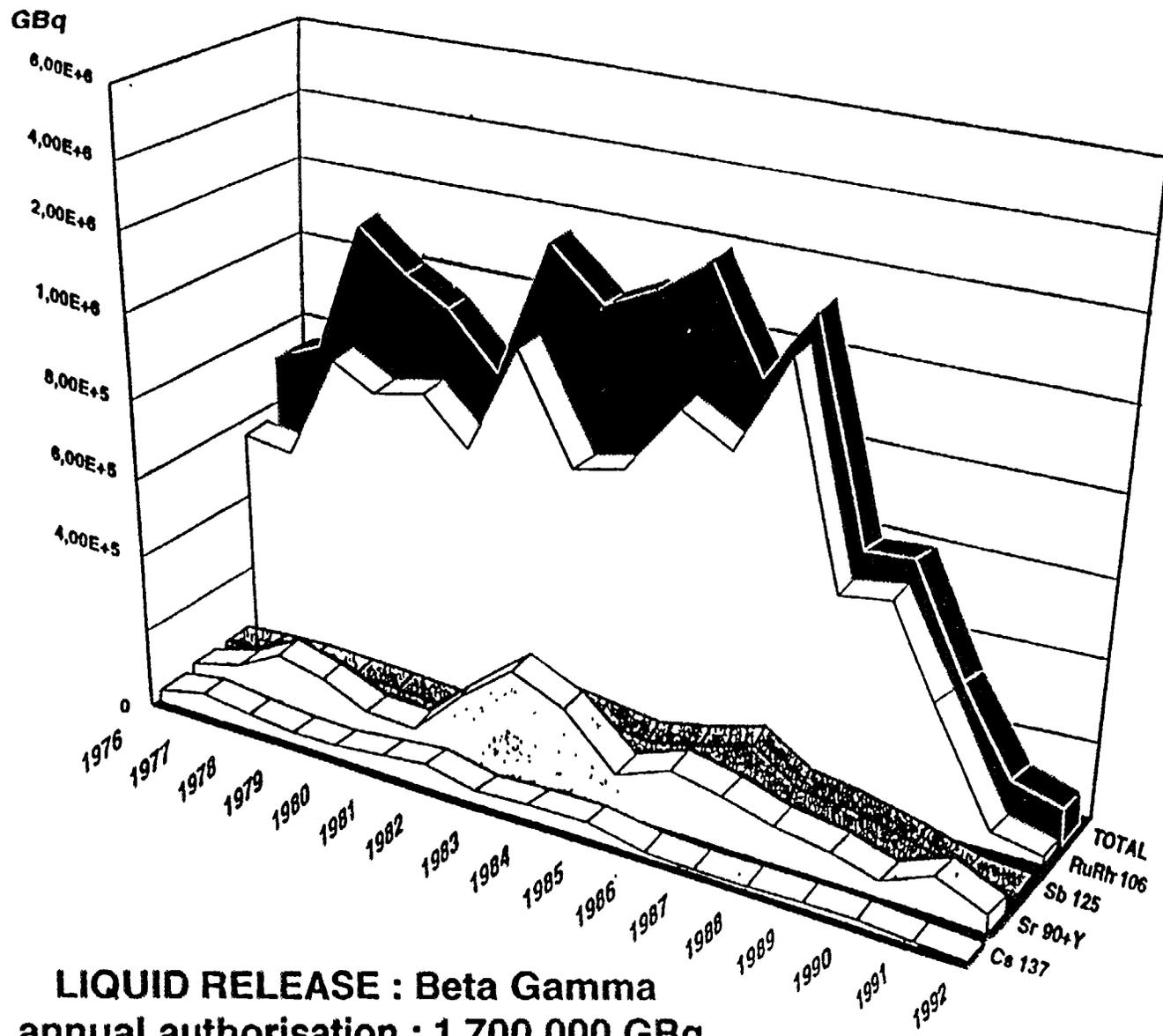


FIG. 2. Liquid released to the sea

Results of Monitoring [3]

The La Hague monitoring programme, which has been certified by the Ministry of Health, is a source of valuable data. Figure 1 shows environmental monitoring data for the last ten years, along with measurements taken in 1965 and 1966, the reference years for natural site conditions prior to start-up of the plant.

The highest activity levels are also the oldest (at the back of the table), and relate to marine monitoring. Activity levels have decreased over the last ten years, while the quantities of reprocessed fuel continued to grow. This decline in activity reflects lower releases of activity to the sea (Figure 2).

Figure 1 also shows that, for the last few years, man-made radioactivity has remained at the same level as natural radioactivity (potassium 40 and beryllium 7) for limpers, and less for other samples.

Conclusion

The annual effective dose of the exposed members of the public may be estimated at about 10 μSv . In comparison, the natural effective dose of the La Hague's population is about 2400 $\mu\text{Sv}/\text{year}$. The very good records displayed by the La Hague reprocessing plant, related to safety, are essential for the health of workers as well as neighbours of the plant; but they also confirm that recycling technology is an established fact.

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HEALTH AND ENVIRONMENTAL IMPACTS OF REPROCESSING IN INDIA

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Abstract

India has at present an operating plant for reprocessing spent fuel located at Tarapur on the west coast of India. The plant has a capacity of 100 t/y of spent fuel. This report gives an example of the evaluation of health and environmental aspects of a reprocessing plant.

Introduction

India has established full-fledged nuclear fuel cycle facilities in support of its civilian nuclear power programme. These are being further expanded in stages with the objective of meeting the additional requirements for the 10,000 MW(e) installed capacity targeted to be achieved by the turn of the century. In view of its limited resources of natural uranium the programme envisages plutonium recycle in thermal as well as fast reactors and the utilization of its vast resources of thorium in ^{233}U fuelled breeders in the long run. Reprocessing and recycle forms, therefore, a key element in India's nuclear power programme.

India has presently an operating plant for reprocessing spent fuel located at Tarapur on the west coast of India approximately 100 km from Bombay. This plant has a capacity of 100 t/y of spent fuel. Another plant with similar capacity is being built at Kalpakkam about 100 km from the city of Madras in south India, and is expected to go into operation in 1996. Both these plants will essentially reprocess spent CANDU natural uranium fuel irradiated to about 6000 Mwd/te and in practice cooled for 3 - 5 years. This cooling time enables the circumventing of a number of process-related and radiological safety problems namely transporting spent fuel in dry condition without any forced cooling, absence of troublesome fission products such as ^{95}Zr - ^{95}Nb and ^{106}Ru , less cooling requirements for the high active liquid waste and the vitrified glass canisters and lower worker and public doses. In India it is preferred to co-locate reprocessing and associated waste management facilities including recycle fuel fabrication plants at the same site as that of a nuclear power station since this approach considerably reduces transportation problems and costs and reduces risks in the public domain.

The reprocessing plant also can thus take advantage of the 1.6 km exclusion zone that is presently mandatory for a nuclear power plant in India. The reprocessing plant also incorporates conversion and calcination facilities for the conversion of uranyl nitrate and plutonium nitrate solutions to their oxide forms. The plant employs a chop-leach head-end and the conventional Purex process with modifications based on experience. The future plants would incorporate redundant lines of production, remote systems technology for maintenance and intervention of process equipment and optimised lay out for servicing and repair.

Environmental safety considerations and impact assessment

Environmental safety has received considerable emphasis and has top priority in the Indian nuclear programme. This is true of reprocessing as well. It must be stressed that in terms of likely accidents and their consequences the reprocessing plant is more benign as compared to a nuclear power reactor on a number of considerations such as considerably lower potential energy available for causing a dispersal of radio-active material, absence of volatile or gaseous fission product species such as ^{131}I and short lived fission gases and larger time constants for energy transients. The operating temperatures and pressures

are also much lower; in fact the process systems are operated under negative pressure. On the other hand the reprocessing plant handles highly reactive and corrosive solutions of high specific activity and presents a different and complex problem of containment as compared to a power reactor although the same principle of defence in depth and multiple barriers is followed in the design and operation of such plants.

Being chemical plants, problems similar to a chemical plant can also exist but the stringent requirements imposed for the containment of radioactivity automatically reduces chemical pollution hazards.

The principal dose limit of 1 mSv/y for a member of the critical group in the public domain is followed in the design and operation of nuclear facilities in India. For co-located facilities this limit is inclusive of future facilities and expansions as well. In practice only 0.5 - 0.8 mSv/y is utilized for planning purposes. This is further apportioned among the various facilities operating and planned at the site, and further among the atmospheric, aquatic and terrestrial routes and also for specific radio-nuclides depending on the technical characteristics of the installation taking into account all significant pathways of exposure. The ALARA principle is also applied in arriving at these figures.

Based on these concepts a three-tier system of regulatory control and compliance is employed for radiological surveillance of effluents and the resulting exposure in the public domain arising from nuclear operations:

- (I) discharge criteria are specified for each plant in the form of Technical Specifications for plant operations
- (ii) all effluents before they are discharged from the plant are sampled and monitored at the source to ensure that discharge criteria are being met and
- (iii) an independent means of monitoring the environment by organizing and conducting a detailed environmental monitoring programme is established.

These detailed measurements are carried out by the Environmental Radiological Laboratory located at each of the main sites in the country and operated by the Health Physics Division of the Bhabha Atomic Research Centre.

The primary aim of the environmental monitoring programme is to demonstrate compliance with the radiation exposure limits set for members of the public. This requires detailed measurements of radio-activity content in a number of environmental matrices. The samples are selected on the basis of potential pathways of exposure. The number and type of samples and sampling frequency can be site-specific depending on the nature of the operations, utilization of the local environment, existence of population clusters etc. A Quality Assurance Programme in which results of selected measurements are compared with international standards such as those of WHO, IRC and the IAEA, ensure the quality of measurement and the data. Although the primary emphasis is on samples that are relevant directly to the estimate of dose such as drinking water, edible food items, air, etc., a number of other samples are also assayed for radio-activity and used as trend indicators and sensitive indicators or markers. Examples of the former are sea water, sediment and the latter, goat's thyroid which concentrates the radio-nuclide ¹³¹I to a great extent, if present in the environment.

Presently a reprocessing plant will be designed and operated so that the exposure for a member of the public will not exceed 0.03 mSv/y. The present experience shows that environmental releases are small and easily controlled to meet the above criteria without any difficulty.

Segregation of liquid effluents and dedicated treatment plants for treating plant effluents such as evaporator condensates, fuel storage pond ion exchange regenerant effluents as well as alternate methods for reducing effluent generation such as disposable cation exchange beds etc. have resulted in very small effluent discharge values for the Indian reprocessing plants. Ten year (1981 - 1990) annual average figures for the reprocessing plant at Tarapur has been as follows:

Liquids:	Alpha: 80 mCi/y (3 GBq/y)	Beta: 4.0 Ci/y (150 GBq/y)
Gaseous:	Alpha: 250 μ Ci/y (10 MBq/y)	Beta: 13.5 mCi/y (0.5 GBq/y)

Studies have also been conducted on the behaviour and release of ³H and ¹²⁹I and to a limited extent ¹⁴C.

The observations are as follows:

- HTO: 25 - 35% of the inventory is released to the environment out of which 20 - 30% is in liquid effluent mainly in the final condensate from the evaporators. The remainder is estimated to be present in zircalloy cladding.
- ¹²⁹I: 10% of the inventory is released out of which 8% is released through stack mostly during dissolution and 2% in liquid effluent.
- ¹⁴C: limited measurements indicate 10% of the inventory is released mostly in gaseous effluent with little in liquid effluent.

The dose to a member of the public in the critical group from the atmospheric and aqueous effluents including Kr⁸⁵, I¹²⁹, HTO, the fission products and the actinides is estimated to be less than 10μSv/y.

Since the Pressurized Heavy Water Reactors also release ¹⁴C and HTO it is seen that the contribution from the associated reprocessing plants is less than that from a nuclear station of this type which is less than 10μSv/y from both atmospheric and aquatic releases.

In so far as non-radiological pollutants are concerned, the emissions must conform to national pollution control standards.

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IMPACT OF THE TOKAI REPROCESSING PLANT ON THE WORKERS AND ON THE SURROUNDING ENVIRONMENT

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Abstract

The Tokai reprocessing plant began operation in September 1977 to establish oxide fuel reprocessing technology in Japan. Its designed capacity is about 0.7 metric tons of uranium per day. This report gives an example of the evaluation of the health and environmental aspects of a reprocessing plant.

1. Introduction

The Tokai reprocessing plant began its operation in September 1977 to establish oxide fuel reprocessing technology in Japan. Its designed capacity is about 0.7 metric tons of uranium per day, and it has been reprocessing 70 - 80 tons per year in recent years, which amounts to 717 tons in total by the end of March 1994.

2. Occupational Exposure

When we measure the occupational exposure of the workers in the reprocessing plant, external exposure by γ -rays and neutrons is usually important in normal operation, but internal exposure by inhalation and/or ingestion of radioactive nuclides must be carefully considered in maintenance works or accidental cases.

According to data collected over the past 5 years, the number of monitored workers is around 2,500 per year and the annual collective effective dose is around 0.5 to 1.5 man Sv, which doesn't depend on the amount of reprocessed fuels but on the kind of maintenance work carried out.

The average annual effective dose per monitored worker is around 0.5 mSv and normalized collective effective dose is around 0.2 man Sv per GWa.

3. Environmental Impact

In the Tokai reprocessing plant, authorized limits for annual discharge are as follows:

Authorized Annual Discharge Limits			
Airborne Effluents (GBq/a)		Liquid effluents (G Bq/a)	
Kr 85	8.9×10^7	Gross β (except for H)	9.6×10^2
H 3	5.6×10^5	H 3	1.9×10^6
C 14	9.7×10^3	Sr 90	3.2×10
I 129	1.7	Cs 137	5.5×10
I 131	1.6×10	I 129	2.7×10
		Pu	2.3

Measured annual discharge in 1993 is as follows:

Measured annual discharge (1993)			
Airborne effluents (G Bq/a)		Liquid effluents (G Bq/a)	
Kr 85	5.2×10^6	H 3	1.6×10^5
H 3	2.1×10^3	Sr 90	2.2×10^{-3}
C 14	3.2×10^2	Cs 137	1.1×10^{-2}
I 129	1.3×10^{-1}	I 129	5.5×10^{-2}
I 131	ND	Pu	3.0×10^{-3}

Annual effective dose for the public around the plant site is estimated for potential pathways with site-specific parameters such as food consumption, concentration factors of marine organisms, and meteorological conditions. Based on the above measured discharge, maximum annual effective dose for the public is estimated to be around $1 \mu\text{Sv}$ per year, which corresponds to 0.1% of the annual dose limit for the public.

Extensive radiation monitoring has been conducted in the environment around the plant site, and no significant environmental effect from the plant operation has been found.

PART III
COUNTRY REPORTS

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**ASSESSMENT OF RADIOLOGICAL AND NON-RADIOLOGICAL
HAZARDS IN THE NUCLEAR FUEL CYCLE - THE INDIAN EXPERIENCE**

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Abstract

Design and operational aspects of nuclear fuel cycle facilities have several features that distinguish them from nuclear power plants. These are related to (i) the nature of operations which are chiefly mining, metallurgical and chemical (ii) the nature and type of radio-active materials handled, their specific activities and inventories and (iii) the physical and chemical processes involved and the associated containment provisions. Generally the radioactive materials are present in an already highly dispersible or mobile form, in the form of solutions, slurries and powders, often associated with a wide variety of reactive and corrosive chemicals. There are further marked differences between the front-end and back-end of the fuel cycle. Whereas the front-end is characterised by the presence of large quantities of low specific activity naturally occurring radioactive materials, the back-end is characterised by high specific activities and concentrations of fission products and actinides. Radioactive characteristics of waste arisings are also different in different phases of the nuclear fuel cycle. Potential for internal exposure in the occupational environment is another distinguishing feature as compared with the more common designs of nuclear power reactors. Potential for accidents, their phenomenology and the resulting consequences are also markedly different in fuel cycle operations. The non-radiological hazards in fuel cycle operations are also of significance, since the operations are mostly mining, metallurgical and chemical in nature. These aspects are examined and evaluated in this paper, based on the Indian experience.

1. INTRODUCTION

India has established full-fledged nuclear fuel cycle facilities in support of its civilian nuclear power programme. These are being further expanded in stages with the objective of meeting the additional requirements for the 10 000 MW(e) installed capacity targeted to be achieved by the turn of the century. In view of its limited resources of natural uranium, the programme envisages plutonium recycle in thermal as well as fast reactors and the utilization of its vast resources of thorium in ^{233}U fuelled breeders in the long run. On account of this, India's nuclear fuel cycle activities and programmes are diverse and wide-ranging. The operations include mining and milling of uranium and thorium ores, production of nuclear grade uranium and thorium oxides, fabrication of fuel elements both for LWRs and PHWRs, spent fuel storage and reprocessing. It has established facilities for the manufacture of mixed-oxide and mixed-carbide fuel elements for plutonium recycle in thermal/fast reactors and has acquired pilot plant experience in the extraction of ^{233}U from irradiated thorium and the manufacture of ^{233}U - Al alloy plate-type fuel elements for a low-power research reactor. In addition, facilities have been established at all sites for the processing, conditioning, handling and disposal of the different categories and types of radioactive wastes that arise in the different phases of the nuclear fuel cycle. With the exception of the long-term disposal of high active waste arising from the back-end of the fuel cycle, the nuclear fuel cycle in India can be considered to be fully operational and constitutes a unique experience among the developing countries.

Simultaneously with the above activities, an on-going programme for the assessment and monitoring of the radiological impact of these diverse operations both in the occupational and public environment has been operating successfully for a number of years and the radiological burden of the various components of the fuel cycle has been assessed in both these domains.

2. QUANTITATIVE EVALUATION OF HEALTH AND ENVIRONMENTAL IMPACTS

Quantitative evaluation of health risks and environmental impacts from radioactivity associated with the different stages of the nuclear fuel cycle will offer much needed insights for (i) comparison of these risks with similar risks in non-nuclear fuel cycle operations for the production of electricity (ii) within the nuclear fuel cycle itself, the contribution of radiological impact to the overall health and environmental impact arising from those aspects not related to radioactivity since most of these operations - mining, metallurgical and chemical, have several features in common with conventional industrial operations and (iii) within the nuclear fuel cycle, considering only the radiological impact, the need for assigning priorities in radiation protection for the different components based on a systems approach and optimization.

In so far as the last mentioned aspect is concerned, it is becoming increasingly clear that mining operations in the front-end of the fuel cycle and the management of low level wastes in all the phases of the nuclear fuel cycle, not necessarily nuclear power reactor operations and high active wastes from the back-end of the fuel cycle, are areas requiring attention in the current state of development in so far as radiological impact from normal operations is concerned, especially in the light of the recent lowering of the occupational exposure limits by ICRP. The simple reason, not so obvious, is that, although the specific activities of the materials handled are small, the quantities involved are large and the concept of containment, shielding etc. that are so readily applied and applicable to other phases of the nuclear fuel cycle where the quantities are far smaller, are not so readily applied in so far as these operations are concerned. Augmented mine ventilation, mechanized and automated material handling systems, better containment provision during fuel manufacture and for low level wastes, are examples of solutions that will require examination. A similar situation is likely to occur in the long-term management of high active and alpha-emitting wastes arising from the back-end of the fuel cycle in resorting to schemes such as recovery of actinides from waste solutions, converting them to solid elements and their subsequent transmutation to short-lived species in fast reactors if the entire scheme is not viewed in its totality and consideration is not given for the risks involved in operating additional plants, generation of secondary effluents and wastes and the need for decommissioning of these plants etc. Adopting a systems approach to safety will also mean that there has to be at least a national consensus on the monetary valuation of health detriment in all fields of human activity lest a totally distorted approach towards risk reduction is applied in a few sectors such as nuclear power resulting in valuable monetary resources becoming unavailable for other vital areas of safety, health care and well-being both of workers and the public and the ALARA philosophy is applied not in its true spirit but used for rationalizing unjustifiable expenditures in the name of 'risk perception' [1].

3. DISTINGUISHING FEATURES OF FUEL CYCLE OPERATIONS

From the point of view of health and environmental impact nuclear fuel cycle operations have features that distinguish them from nuclear power plants both from the point of view of normal operations and accidents. These are related to (i) the nature of operations which are chiefly mining, milling, metallurgical and chemical (ii) the nature and type of radioactive materials handled, their specific activities and inventories and (iii) the physical and chemical processes involved and the associated containment provisions.

Unlike in nuclear reactors, generally the radioactive materials are present in an already highly dispersible or mobile form, in the form of solutions, slurries and powders often associated with a wide variety of reactive and corrosive chemicals. There are further marked differences between the front-end and the back-end of the fuel cycle. Whereas the front-end is characterised by the presence of large quantities of low specific activity naturally occurring radioactive materials, the back-end is characterised by high specific activities and concentrations of fission-products and actinides. Further in the normal operation of the reactors most of the activity is well contained within the fuel element and the radiation hazards mainly arise from the activation products and the small quantities of fission products that leak from the reactor core into the primary coolant. In a fuel reprocessing plant however, these are present in very high concentrations along with actinides often in the presence of highly corrosive and reactive chemicals [2, 3]. In the waste vitrification plant, the concentrated fission product solutions are again subjected to high

TABLE I Radiological and Non-Radiological Hazards in the Nuclear Fuel Cycle

Fuel Cycle Activity	Occupational		Environmental	
	Radiological	Non-radiological	Radiological	Non-radiological
1. Mining & Milling				
(i) Uranium ore	Ext: not significant exp: Int: Significant exp: ^{222}Rn	Silica dust Heat Noise Vibration	^{222}Rn from mine exhaust and tailings pond U, ^{230}Th , ^{226}Ra , ^{210}Po in aquatic releases	SO_2 - atmospheric Mn, SO_4 , chlorides in aquatic releases
(ii) Thorium ores	Ext: Significant Int: ^{220}Rn ^{228}Ra , Th	Silica dust NaOH	^{228}Ra , in aquatic releases	P, Po_4 , Pb in aquatic releases
2. Fuel Fabrication				
(i) Zirconium plants	Nil	NO_2 , HF, Cl_2 , HCl Cl_2 , $\text{NO}_2\text{H}_2\text{SO}_4$		Fluorides, Nitrates
(ii) Fuel plants	Ext: Negligible Int: UO_2	NO_2 , NH_3 , HF	Insignificant	Chemical effluents containing sodium nitrate, sodium silicate, ammonium nitrate, ammonium sulphate, sodium fluorides are generated in significant quantities

TABLE I (cont.)

Fuel Cycle Activity	Occupational		Environmental	
	Radiological	Non-radiological	Radiological	Non-radiological
3. Spent Fuel Reprocessing and high active waste immobilization	Ext: Significant Int: ^{239}Pu	NO_2 , hydrazine, formaldehyde TBP/Kerosene, ammonia, styrene	^{85}Kr , ^3H ^{129}I , ^{106}Ru , ^{137}Cs Actinides	NO_2 , NH_3 TBP
4. MOX/MC fuel fabrication	Ext: neutron/soft X-ray gamma exposures from high burnup fuel plutonium Int: Plutonium		Pu-atmospheric long-lived actinide bearing solid waste	
5. Heavy Water Plants	Nil	NH_3 , H_2S SO_2		H_2S SO_2 NH_3

TABLE II Reactors and spent fuel reprocessing plants - Relative Hazards

Feature	Reactor	Reprocessing Plant
Inventory of long-lived fission products in dispersible form	Low	High
Inventory of volatile/gaseous radio-active species	Very high	Orders of magnitude less
Specific activity of solutions handled	$\mu\text{Ci/ml}$	Ci/ml
Medium	Water	reactive and corrosive chemicals, acids, alkalis, organic solvents
Access to radio-active material/availability	Not very significant	Significant
Problems with long-lived alpha emitters	None	Significant
Potential for skin exposure	Little	Significant
Potential for internal exposure	Not significant (except in PHWRs)	Significant
Operating conditions	High temperature and pressures	Low temperature, operations generally under vacuum
Potential energy available for dispersal of radio-activity	Very high	Low
Time constants for energy release transients	Low	High
Dispersal mechanisms	Power excursion, loss of coolant results in sudden overheating of fuel	Fires, exothermic chemical reaction involving reactive chemicals/flammable substances, pyrophoric powders Criticality incidents involving fissile solutions Loss of cooling to high active liquid wastes

TABLE III Potential accident scenarios in fuel cycle facilities

Facility	Radiological	Non-radiological
Mining and milling	--	Mine fires, explosions, flooding, conventional mining accidents
Fuel fabrication		Fires involving pyrophoric materials (e.g. Zirconium sponge, scrap, magnesium); Toxic gas release
Spent fuel reprocessing	<p>Exothermic run-away chemical reactions, during dissolution, evaporation or conditioning of process solutions</p> <p>Fires involving loaded solvents/ion exchange resins</p> <p>Cask drops, dropping of other heavy loads in spent fuel storage bay</p> <p>Loss of cooling to high active liquid waste storage tanks</p> <p>Criticality excursions</p> <p>Fires in plutonium powder handling facilities</p>	Conventional fires involving solvents and diluents, or other reactive chemicals
Waste immobilization	<ul style="list-style-type: none"> - Fires during bitumenisation - Runaway chemical reactions during polymerization - Waste canister drops; active glass spillage 	
Waste storage	<p>Ground water incursion</p> <p>Natural calamities</p> <p>Human intrusion</p>	

temperature processes and converted to a solid form suitable for ultimate disposal. In the mixed oxide/mixed carbide fuel fabrication plants the problem is one of handling large quantities of highly toxic and pyrophoric powders [4]. Although the larger inventory of the short lived fission product noble gases and volatile species such as iodines are largely absent, certain other long lived species such as ^{14}C , ^{129}I , ^3H and ^{85}Kr and the actinides make their appearance and become available. Radioactive characteristics of waste arisings are also different in different phases of the nuclear fuel cycle. Potential for internal exposure in the occupational environment to radon and thoron daughters as well as to the long-lived components such as uranium, radium and thorium in the front-end of the fuel cycle and the long lived actinides in the back-end is another distinguishing feature when compared with the more common designs of nuclear power reactors.

Table I provides a summary of radiological and non-radiological hazards that prevail in the different phases of the nuclear fuel cycle from mining and milling of uranium and thorium ores to spent fuel reprocessing. It can be seen that chemical pollutants present significant health risks both to occupational workers and in the public environment, unless careful attention is paid for their control and abatement. This is an aspect that distinguishes most fuel cycle operations from nuclear power plants.

Table II brings out those features in the design and operation of a spent fuel reprocessing plant having an influence on health and environmental impact assessment as compared with nuclear power reactors.

Table III provides a brief summary of accident scenarios that can be considered credible in various fuel cycle operations. Generally the consequences in the off-site environment are minimal and impact if any, will be confined to plant premises. This has been borne out by experience.

While dealing with probable events with potential for exposure, it will be advantageous to adopt common criteria for all the phases of the nuclear fuel cycle, inclusive of power reactor operation. Indeed, in the context of acceptable risks from accidents, it would be necessary to arrive at and adopt common criteria for all types of accidents - chemical, transportation, mining and metallurgical operations, manufacturing etc. in order to ensure common approach towards risk reduction in all forms of industrial activity.

4. RADIOLOGICAL IMPACT OF THE NUCLEAR FUEL CYCLE - THE INDIAN EXPERIENCE

The following sections deal with an assessment of the radiological impact of the different operations in the nuclear fuel cycle based on operational experience in India.

4.1. Uranium mining and milling

4.1.1. Occupational exposure

In India, the major uranium mine is an underground mine located at Jaduguda in the State of Bihar. The ore is of low grade, about 0.04%. Data for external radiation exposure and internal exposure from radon and its daughters are given in Tables IV, V. Based on the uranium requirement of 152 Tg per Gw(e) - y for a Pressurized Heavy Water Reactor (PHWR) the total collective dose from both mining and milling works out to 19.5 person - Sv per Gw(e) - y out of which 17% is from milling and extraction and the overall internal dose contribution from radon and daughters is 82%. Internal dose contribution from ore dust is negligible in view of the low grade of ore [5].

4.1.2. Exposure to members of the public

Exposure to members of the public from uranium mining and milling arises from three sources namely:

- i) inhalation of radon daughters from mine and mill exhaust
- ii) radon from tailings pond
- iii) ingestion dose due to ^{226}Ra

TABLE IV Occupational exposure in uranium mining - Jaduguda underground mine

Year	Annual amount of uranium extracted (t)	Annual collective effective dose (person-Sv)	Average annual effective dose per worker (mSv)
1981	124	22.8	20.8
1982	121	19.2	17.5
1983	128	22.5	20.5
1984	140	24.0	17.8
1985	130	25.1	18.6
1986	144	28.5	21.1
1987	141	25.2	18.7
1988	173	25.6	19.0
1989	163	17.7	13.1
1990	157	19.1	13.8

TABLE V Occupational exposure for uranium milling and extraction - Jaduguda

Year	Annual amount of uranium extracted (t)	Annual collective effective dose (person-Sv)	Average annual effective dose per worker (mSv)
1981	124	4.9	10.8
1982	121	5.0	10.0
1983	128	5.1	10.2
1984	140	5.1	10.2
1985	130	5.2	10.3
1986	144	4.2	7.0
1987	141	6.4	10.6
1988	173	4.1	6.9
1989	163	3.9	6.5
1990	157	3.1	5.4

Using the UNSCEAR model for estimating the collective dose due to inhalation of radon daughters from mining and milling and demographic data for India, the collective dose up to 2000 km radius is estimated as 1.8 person Sv per Gw(e)-y. Radon from tailings pond comes mainly from the long-lived ^{230}Th in addition to the small quantities of unrecovered uranium. Using parameters such as uranium content of ore 50% of which is used as back-fill, recovery efficiency and the utilisation of about 1 ha per Gw(e)-y for the tailings pond, the radon emanation rate considering the nature of the top cover and the truncated collective effective dose equivalent commitment model of UNSCEAR, a value of 0.7 person Sv per Gw(e)-y is obtained. The total dose to a member of the public has therefore been estimated as 2.5 person Sv per Gw(e)-y, the dose integration being done over a period of 70 years [6].

Liquid effluents from both the mine and the mill are substantially less important in the context of public exposure. Dose due to ingestion of ^{226}Ra is very small and is hence not considered. The pathway identified is soil-grass-milk and the transfer factors are very small. Proposed topping of tailings pond with added vegetation cover is expected to reduce exposures further.

4.2 Monazite separation and thorium extraction

4.2.1. Occupational exposure

The mineral deposits in the West coast of India in the States of Kerala and Tamilnadu are extensively exploited for recovery of monazite such as illeminite, rutile, zircon etc. The monazite is further chemically processed to extract thorium and rare-earths. The major monazite recovery plant is located at Manavalakurichi in Tamilnadu. Individual external exposures vary, depending on whether the worker lives and works in a high background or low background area or vice versa and varies from 1 mSv/y to 3.5 mSv/y. The collective dose is estimated as 0.2 person-Sv per 1000 t of monazite produced. Inhalation hazards are significant only in some operations such as screening, air and wind tables where localised concentration up to 50% DAC are sometimes seen to occur [7].

The monazite sand is further chemically treated at the Rare Earths Plant at Udyogamandal, Alwaye. The per capita exposure per worker from external exposure as well as internal exposure is estimated 13 mSv/y and the collective dose at about 1 person-Sv per 1000 t of monazite processed for recovery of thorium. The total collective dose is estimated as approximately 0.12 person-Sv per tonne of thorium concentrate produced.

4.2.2. Public exposure

With respect to environmental impact, chemical processing of monazite gives rise to liquid effluents containing low levels of ^{228}Ra and phosphates and fluorides. Treatment of these effluents with lime and calcium chloride has resulted in very low concentrations in the effluent receiving water body. The exposure resulting from this source is estimated as $0.5 \mu\text{Sv/y}$ [9].

4.3 Fuel fabrication

Fuel fabrication facilities are located at Hyderabad in the State of Andhra Pradesh. Occupational exposure has been estimated as 0.75 person-Sv per Gw(e)-y. The main hazard in this operation arises from potential inhalation exposure to uranium oxide dust and chemical pollutants. These are however controlled by ventilation and use of protective equipment. Collective dose to the public has been found to be negligible. Effluents contain chiefly chemical pollutants that need careful handling and treatment.

4.4 Spent fuel reprocessing

Data on occupational exposures and environmental releases have been obtained from the Power Reactor Fuel Reprocessing Plant located at Tarapur, having a design throughput of 100 Te/y. It has so far reprocessed PHWR spent fuel both from the Rajasthan Atomic Power Station and the Madras Atomic Power station with an average irradiation of about 6000 Mwd/t and cooling time of 3-5 years. The experience with respect to occupational exposures and environmental releases has been very satisfactory as seen from Tables VI, VII. The occupational exposure for higher capacity plants is estimated at not more than 10 person - Sv per Gw(e)-y. The total contribution from effluents, both liquid and gaseous is estimated as 0.08 person - Sv per Gw(e)-y.

One of the important parameters that influences safety and technical options in the different phases of the back-end of the fuel cycle is the 'cooling time' of the spent fuel prior to its reprocessing for the recovery of fissile material. The advantages of allowing a reasonably long cooling time for spent fuel are:

- a) Safer and less expensive transportation of spent fuel on account of lesser shielding and heat dissipation requirements per unit weight of fuel transported.
- b) Elimination or minimization of troublesome fission products like ruthenium and zirconium during reprocessing from the point of view of decontamination efficiency in solvent extraction cycles.

TABLE VI Collective effective dose and per capita radiation exposure at Power Reactor Fuel Reprocessing Plant (PREFRE), Tarapur

Year	Collective dose Equivalent (person-Sv)	Per Capita exposure (mSv)
1981	1.15	2.62
1982	1.17	2.66
1983	2.10	4.86
1984	2.62	6.07
1985	3.50	7.33
1986	3.17	7.52
1987	2.26	5.43
1988	2.44	6.05
1989	1.62	4.02
1990	1.26	3.10
1991	3.41	8.68

Table VII Environmental Releases from Power Reactor Fuel Reprocessing Plant (PREFRE), Tarapur

A. Liquid Effluent

Year	Alpha activity (mCi)	Beta activity (Ci)
1982	15.7	2.7
1983	121.2	2.9
1984	188.7	6.5
1985	147.4	8.2
1986	67.4	5.8
1987	71.3	2.2
1988	106.7	3.9
1989	43.6	3.0
1990	98.4	4.0
1991	8.5	1.1

B. Atmospheric Releases of particulate activity

Year	Alpha activity (mCi)	Beta activity (mCi)
1982	0.2	2.0
1983	0.4	6.2
1984	0.05	0.6
1985	0.1	1.4
1986	0.3	10.5
1987	0.2	5.6
1988	0.7	76.5
1989	0.6	28.0
1990	0.07	4.0
1991	0.05	0.7

Note: Operations include conversion of plutonium nitrate to oxide

- c) Possibility of direct vitrification of high active waste with minimum interim storage in liquid form.
- d) Absence of volatile RuO_4 in the waste vitrification process resulting in less complex off-gas clean up systems.
- e) Less stringent requirements for interim storage of vitrified waste (air cooling in place of water cooling).
- f) Reduced shielding and man-rem commitments in reprocessing and vitrification plants.

Against these potential benefits must be considered the necessity for treating the increased volume of effluents that will result from longer storage of spent fuel. But this problem can be controlled by storing spent fuel in leak-tight containers in the fuel storage pool.

Based on the experience gained so far, it would appear that the following factors are extremely important for a safe and efficient reprocessing technology:

- a) Reliable chemical plant equipment with extremely low failure rates, requiring almost no maintenance.
- b) Development of a remote systems technology that will enable both in-service inspection and surveillance of radio-active equipment and their maintenance with minimum direct personal intervention.
- c) Provision of redundant lines of production.

Large-scale reprocessing plants of the future would be expected to have also the following design features or provisions [10,11]:

- a) Provision for the removal, handling and disposal of solid fines or residues from dissolver solutions.
- b) Provision for recycling waste evaporator condensate to limit discharge of tritium to the environment.
- c) Systems for the removal of ^{85}Kr and iodines from the dissolver off-gas.
- d) Provision for separation of actinides (^{241}Am) from raffinate waste.
- e) Design for decommissioning, especially the high-active liquid waste storage tanks.

Measurements have been carried out on the releases of long-lived radioactive species such as ^{129}I , ^{14}C and HTO during spent fuel reprocessing. Out of the total inventory of ^{129}I in the fuel 10% is released in low level effluents with 8% appearing in the gaseous effluents, mostly during dissolution of the fuel and 2% in the low level liquid effluents. The plant employed only simple water scrubbing for the off-gas. With regard to tritium, it is observed that 25-35% is released to the environment with the major fraction of 20-30% appearing in the low-level liquid effluents, mainly, the final evaporator condensate. In so far as ^{14}C is concerned limited measurements indicate a release of 10% of the inventory in the gaseous effluent, with no ^{14}C detectable in the liquid effluent.

4.5 Waste management

Definitive data on occupational exposures from waste management plants related to the back-end of the fuel cycle are not yet available. But data from the operating waste management plants indicate that their contribution is unlikely to be significant. Data available with respect to shallow land disposal of low-active

TABLE VIII Collective and per capita doses estimated for nuclear fuel cycle operations in India

	Occupational collective doses person-Sv per GW(e)-yr	Doses to members of the public person-Sv per GW(e)-yr
Uranium mining and milling	19.5	--
Radon from mines		1.8
Radon from tailings pond		0.7
Total to public		2.5
Fuel fabrication	0.75	Insignificant
Spent fuel reprocessing	10.0	0.08

TABLE IX Steps taken to reduce environmental impact of fuel cycle operations in India

Uranium mining and milling	Effluent from tailings pond as well as those arising from mining and milling operations are treated with BaCl ₂ and lime to reduce ²²⁶ Ra and manganese level to low values. An effluent treatment plant has been in operation for this purpose. Topping of tailings pond and added vegetation cover proposed for tailing pond area.
Thorium extraction from monazite	An effluent treatment plant has been in operation since 1981 for the removal and retention of ²²⁸ Ra, Po ⁴ and F present in the effluents by treatment with lime and calcium chloride.
Fuel fabrication	Liquid effluent contains chiefly sodium nitrate, sodium silicate, ammonium nitrate, ammonium sulphate, sodium fluoride etc. These are handled in lined solar evaporation ponds for recovery of salts. Electrostatic precipitators for control of uranium dust emission and scrubbers for recovery of Cl ₂ , NO ₂ etc. from gaseous effluents reduce pollutant levels in the environment well below prescribed standards.
Spent fuel reprocessing	Treatment of spent fuel storage bay water with non-regenerable resins, recycling of final evaporator condensate.

wastes indicate negligible exposure to the public from these operations. No ground water contamination has occurred so far in any of the sites, as a result of these facilities.

4.6 Radiological impact for the nuclear fuel cycle

Table VIII summarises collective and per capita doses presently estimated for fuel cycle facilities in India. The Indian experience indicates that, as elsewhere in the world, uranium mining and milling is a component of the fuel cycle which will require further attention from among the various fuel cycle activities from the point of view of minimising the radiological impact both in the occupational and public domain. Conventional hazards in uranium mining are also significant. Some of the steps taken to reduce environmental impact from fuel cycle operations in India over the last few years are presented in Table IX.

4.7 Non-radiological impacts in the nuclear fuel cycle

Health and environmental impacts arising from chemical pollutants in the work environment as well as in the public domain need to be quantified. Although concentrations of significant pollutants are being routinely assessed and are well below prescribed industrial hygiene standards, these data cannot be easily translated into health effects quantitatively. This is an area for further investigation.

Mention must be made here of the heavy water production plants that form an integral component of the nuclear fuel cycle operations in India. Heavy water is the moderator and coolant and is an essential input for a natural uranium fuelled Pressurized Heavy Water Reactor (PHWR), the mainstay of the first stage of the Indian nuclear power programme. PHWRs require an initial inventory of about one tonne of heavy water per MW(e) of installed capacity. The replenishment requirement is about 10% per year to make up for losses. To meet this requirement, eight heavy water plants have been commissioned and are operational in different parts of the country. The processes employed are chemical exchange between (i) hydrogen sulphide and water and (ii) ammonia and hydrogen. These plants have large inventories of these toxic and flammable gases and operate under very high pressures and carry significant hazard potential.

Data are available on occupational fatalities and lost-time accidental injuries in fuel cycle facilities for the period 1971-1991. These are presented in Table X [12]. A scrutiny of the data enables a reasonable comparison with radiological risks. It is observed that (i) fatality rate from conventional accidents works

TABLE X Facilities in fuel cycle operations in India (1971 - 1991)

Activity	No. of Fatalities
Mining and milling	14
Fuel fabrication	4
Heavy water production	5
Spent fuel reprocessing	1
Total	24

Note: The above works out to an actual fatality rate of 1.2 per year; if one adds the contribution of lost-time accidental injuries based on the severity rate, the effective equivalent fatality rate works out to 1.8 per year.

out to 1.8 per year, taking into account lost-time accidental injuries also based on their severity rate whereas the projected cancer fatality risk is about one per year based on the annual collective effective dose of 20-25 person-Sv and applying the most recent ICRP-90 risk factors for cancer (ii) the main contribution to both these components comes from uranium mining highlighting the fact that this is the most important area where investment for safety improvements are called for and (iii) the rest of the fuel cycle activities - most notably spent fuel reprocessing appear to be of lesser concern.

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ENVIRONMENTAL IMPACT OF THE NUCLEAR INDUSTRY IN CHINA

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Abstract

Since its foundation in 1955, the nuclear industry has become a comprehensive industrial, scientific and technical system in China. The nuclear industry has obviously brought great profit to the country, but how much environmental effect it has caused is a question of common interest which we should answer. This report shows the environmental assessment of the nuclear fuel cycle in China.

1. PURPOSE OF THE ASSESSMENT

At present, the nuclear industry has entered a new development period, i.e. the period of exploration of nuclear energy and wide use of nuclear technology and radioactive isotopes. The 300 MW(e) unit of Qinshan Nuclear Power Plant has reached full power capacity and the Daya Bay Nuclear Power Plant 900 MW(e) unit x 2 are under construction. The two units with 600 MW(e) installed capacity of the Qinshan Nuclear Power Plant have been sited. District heating reactors and reactors for both heating and electricity are on the way to be built. Nuclear Power Plants are different from other nuclear facilities, for they need to be located near a city or cities. Reactors supplying heat and both heat and electricity must be built in a city or near cities. Nuclear technology and radioactive isotopes are utilized increasingly in medicine, industry, agriculture, and scientific research, and have entered into our daily life. Therefore, the public show more and more concern for the safety of nuclear practices and their potential impact on the environment. The history of the world nuclear energy and nuclear industry shows that nuclear energy is a clean one with its good safety record, which is unchangeable even by such a severe accident as the Chernobyl accident. This has been proved by plenty of research achievements in environmental assessment of energy sources in other countries. Is this conclusion consistent with the specific cases in China? The assessment of the environmental quality of the current nuclear industry in China will be helpful to answer it, provide a basis for the environmental assessment of the nuclear energy in China, and be of benefit to the environmental assessment of energy sources in China as well.

It is necessary to assess the environment quality systematically so as to scientifically and quantitatively analyse and study the current situation of the nuclear industry environmental protection and to find out the problems to be solved as well as the solutions to them, for example, to investigate which nuclear system has the biggest effect on the environment in the nuclear fuel cycle, to define the critical nuclide and the critical exposure pathway to the population, to make sure the effectiveness of the present waste treatment, disposal and management, to make clear whether the distribution of environmental protection investment has achieved the best result, and so forth. On the basis of an all-round assessment of environment quality, the best way to improve the nuclear industry environment quality can be determined according to the principle of radiation protection optimization.

The work of environmental assessment began in 1981. The radiological environmental assessment was completed in 1989. The non-radiological environmental assessment began in 1990, will be finished in 1993.

2. RADIOLOGICAL IMPACT

2.1. Methods and models of environmental assessment

2.1.1. Assessment methods

For the sake of the quantitative estimation of the environmental impact due to the 30 year operation of the nuclear industry in China and the comparison between home and abroad, the assessment areas are unified as a round area of 80 km in radius, the center of which is set at the main release point. The distances are 1, 2, 3, 5, 10, 20, 30, 40, 50, 60, 70 and 80 km respectively. The population in the assessment areas is divided into 3 age groups for internal exposure estimation: infant, 0 to 6 years old; juvenile, 7 to 18 years old; adult, older than 18 years old.

2.1.2. Assessment models and parameters

After an intensive investigation and study of the available models and parameters home and abroad, one set of common models and parameters are recommended for "Radiological Environment Quality Assessment of the Nuclear Industry in China over the past 30 Years", according to the features of nuclear industry in China. The main references are Safety Series No. 57 of the IAEA and Regulatory Guide 1.109 of US NRC.

Straight trajectory Gaussian model is applied in the case of airborne release. Atmospheric stability classification is conducted uniformly by P-T method. Combining with a number of large scale atmospheric dispersion experiments, different dispersion parameters are selected for each facility in accordance with its specific topographical characteristics: P-G curves for plain area, data based on the measurements in Julich and Germany for hilly regions, and Briggs' city parameters for mountainous and urban areas. In the calculation of annual concentration in air and surface deposition rate of radioactive nuclide of each subsection of the area of interest, plume depletion caused by dry deposition, wet deposition and nuclide decay are considered, and, meanwhile, correction is made for the influence of plume rise, calm condition, mixture height, valley width and building wake.

In the calculation of radioactive liquids in rivers, an analytic solution of two dimensional convection and diffusion equations is applied, in which only convectational transport and diffusion are taken into account. Based on several dispersion experiments, it is assumed that uniform distribution would be achieved at the down water distance of 40 times of the river width.

Radionuclide concentrations in agricultural products are calculated using the linear transfer model of ecosystem, in which the transfer parameters of nuclide are taken from Safety series No.57 of IAEA, Regulatory Guide 1.109 and technical report NUREG/CR3160 of US NRC. Based upon the above three literatures and the analysis and comparison between the investigation data of some of the facilities, the parameters in the model are recommended for the calculation of nuclide concentrations in crops and animal products.

The following exposure pathways are considered in dose estimation; immersion exposure in plume, immersion external exposure in water, inhalation, external exposure from surface deposition, external exposure from beach deposition and ingestion of contaminated foodstuffs and water. A simple infinite cloud method is used to deal with external exposure due to routine release. concentration factor method is taken to calculate internal exposure to human resulting from food chain transfer. Internal exposure due to inhalation of polluted air is the product of air concentration, breathing rate and dose conversion factor. Individual food and water consumption by each age group is the combination of the data from relevant organizations (e.g. the National Statistics Bureau) and special investigation. The internal exposure dose conversion factors used in the dose estimation are derived upon the principles in Publication 30 of ICRP.

In the dose estimation of accidental release of radioactive material, finite plume model is applied to estimating plume immersion external exposure. In the calculation of deposit external exposure and ingestion exposure through land food chain, it was considered that radioactive materials accidentally released might deposit and then remain on the ground for a long time and continuously result in exposure to the public.

2.1.3. Test of the models and the computerized code

Tests and examinations of the models and their related computer programs for routine release of airborne and liquid radioactive effluent have been conducted to study their suitability. With the basic data (meteorological data, population distribution, food spectrum and so on) of the same facility, comparison was made between computerized codes Y3001 and AIRDOS-EPA. The former was used in the radiological environmental quality assessment in China over the past 30 years and the latter was developed by Oak Ridge National Laboratory of America. Only those nuclides released from the nuclear industry that have bigger impact on the environment are taken into account, for instance, I-131, Rn-222, U-234 and H-3. It is illustrated by the calculation results that the individual dose due to I-131 and U-234 of the two codes mentioned above are consistent within 10%; the dose of 222-Rn computed by AIRDOS-EPA is twice as much as that by Y3001 but is 0.5 in the latter. The doses of H-3 from the two codes are different of times, because the parameters chosen are different significantly. As for the dose estimation models for routine radioactive liquid effluent, comparison between manual calculation and computer calculation was conducted, and the results are the same completely.

2.2. Sites situation

Most of the large nuclear industry facilities are situated in the northwest, southwest and middle-south regions of China. Great attention was paid to the basic requirement of environmental protection and the characteristics of nuclear facilities when siting, and they were located in areas with lower population density or remote regions if possible. The earliest nuclear facilities were set up in rural areas or areas near to mountains that have low population density, less precipitation, low groundwater level, and less land-use. The second set of nuclear facilities were constructed in mountainous or hilly areas, where precipitation is heavy, surface water is very abundant, and ground water table is higher. From the point of view of environmental protection, the sites of these facilities are disadvantageous to some extent, that is a problem of historical cause. Generally, the population density is relatively high in the areas around these facilities, but relatively low in the close vicinity of them. The most of uranium mines are naturally located in the southeast of China, where the population is dense.

The distribution of average population density within 80 km around a facility is shown in Table I for all the facilities of the nuclear industrial systems. The population density distribution around all the nuclear facilities, except mining and research facilities, is shown in Table II. Table III is the average population density for every system. The average population density for whole nuclear industry system is 172 persons per square kilometer and would decrease to 137 persons per square kilometer without considering uranium mining and milling facilities and research facilities. From Table I to III, it can be seen that the population density of the research institutes is the highest, and then in a decreasing order comes the fuel element fabrication plants, uranium mining and milling plants, uranium isotope separation plants, nuclear metallurgical and processing plants.

2.3. Source term: the radioactive materials released to the environment

The radioactive materials routinely released to the environment are those discharged to the environment under the controlled conditions. The annual average release amount, together with their variation range, from the following industrial systems; uranium mining and milling, isotope separation, fuel element fabrication, reactor, reprocessing are shown in Table IV to Table IX.

TABLE I The Population density distribution in the vicinity of all nuclear facilities to the extent of 80 km (man/km²)

Population density	< 10	10-25	>25-50	>50-100	>100-200	>200-300	>300-400	>400-500
Distribution %	2.3	18.6	9.3	2.3	34.9	9.3	14.0	9.3

TABLE II The Population density distribution in the vicinity of all facilities except uranium mining and milling and research institutes (man per kilometer)

Population density	10-25	>25-50	>50-100	>100-200	>200-300	>300-400
Distribution %	18.2	9.1	9.1	45.4	0	18.2

TABLE III The average population density for different kinds of nuclear facilities (man/km²)

Type of facility	Mining	Fuel Fabrication	U Isotopes Separation	Reactor and Reprocessing	Nuclear Metallurgical and Processing	Research Institutes
Population density	160	245	149	66	87	350

TABLE IV Annual release of radionuclides from uranium mining and milling facilities (Bq)

Pathway		Nuclide					
		Rn-222	Ra-226	Th-230	Pb-210	Po-210	U
Liquid	Average	2.76E14	3.94E8	3.68E8	3.68E8	3.72E8	3.58E9
	Range	7.30E10-5.90E14	2/28E5-1.13E9	2.28E5-1.09E9	2.28E5-1.09E9	2.28E5-1.10E9	4.56E5-8.90E9
	Average	---	7.09E10	1.22E10	1.85E10	1.01E10	1.91E11
	Range	---	5.27E7-1.54E11	3.58E7-4.31E10	4.39E7-6.00E10	1.76E7-3.29E10	1.30E8-3.46E11

TABLE V Annual average releases of uranium from uranium isotope separation plants

Nuclide	Pathway			
	Gaseous (Bq)		Liquid (Bq)	
	Average	Range	Average	Range
Total uranium	1.76E9	1.20E8 -2.08E10	5.62E9	4.88E7 -1.37E10

TABLE VI Annual average releases of uranium from fuel element fabrication plants

Nuclide	Pathway			
	Gaseous (Bq)		Liquid (Bq)	
	Average	Range	Average	Range
Total uranium	6.70E9	3.00E5 -1.57E10	8.38E9	6.90E8 -2.91E10

TABLE VII Annual average releases of nuclides from reactors (Bq)

Pathway		Nuclide				
		Ar-41	Xe-133	I-131	Cs-137	Sr-90
Gaseous	Average	1.24E15	3.99E15	7.94E10	---	---
	Range	1.10E14 -2.32E15	2.00E12 -2.82E16	4.00E07 -2.87E11	---	---
Liquid	Average	---	---	---	2.80E08	4.70E08
	Range	---	---	---	1.00E07 -7.00E08	2.00E07 -1.80E09

TABLE VIII Annual average releases of radionuclides from reprocessing plants (Bq)

Pathway		Nuclide			
		Kr-85	Cs-137	Sr-90	Pu-239
Gaseous	Average	4.80E15	1.26E10	2.63E10	1.99E09
	Range	1.44E15 -7.10E15	1.90E09 -4.49E10	3.90E09 -1.56E11	3.40E08 -5.92E09
Liquid	Average	---	5.76E10	8.94E10	9.80E09
	Range	---	2.00E08 -3.70E11	2.30E08 -5.14E11	1.30E09 -6.51E10

TABLE IX Annual average releases of radionuclides from institute of atomic energy (Bq)

Pathway		Nuclide									
		Sr-90	I-131	Cs-137	Po-210	H-3	Pu-239	Xe-133	Ar-41	Ru-106	I-25
Gaseous	Average	4.20E7	1.20E11	4.20E07	2.00E07	1.30E13	1.50E04	5.40E11	1.20E14	4.20E07	2.20E10
	Range	5.30E04 -3.40E08	5.60E08 -9.90E11	5.30E06 -3.40E08	6.70E05 -1.30E08	7.40E10 -1.20E14	1.20E02 -2.40E05	1.20E11 -1.55E13	4.10E12 -5.00E14	5.30E04 -3.40E08	8.40E09 -1.00E14
Liquid	Average	---	---	---	3.10E07	9.16E11	---	---	---	---	---
	Range	---	---	---	2.40E07 -3.80E07	3.30E10 -2.00E12	---	---	---	---	---

The amount of gaseous radioactive materials released to the environment was obtained by multiplying the monitored radionuclide concentration in the effluent with the discharge volume which was actually measured in most facilities and was calculated according to ventilation rates and design data for the very few other facilities. The escape rate of radon from tailing dam is acquired based on the measured escape rate and wind speed. The releases of radioactive materials from natural-evaporating pool is derived on the radioactive aerosol concentration above the pool and wind speed.

The method of calculating the amount of liquid materials released to the environment is nearly the same as that for gaseous materials. The releases of liquid radioactive materials are divided into planned releases and unplanned releases. Planned release of liquid radioactive waste implies the release of the water that is projected to be polluted upon the design basis. In all the facilities except some of the uranium mining, before being released to the environment, the waste water was properly treated and measured to assure that the radioactive concentration was lower than the authorized limits. Therefore, this kind of releases can be determined accurately. Unplanned releases of radioactive water means the releases of radioactive liquid that is not anticipated to be polluted according to the design of the installations, for example, the radioactive liquid discharged through an industrial drainage system or a living drainage system. There was not any requirement for this kind of release in the earlier environmental monitoring program. Compared with planned releases the data of unplanned releases is incomplete.

Quite a lot of radioactive waste water was released into the environment in an unplanned way. Although the radioactive concentration was rather low in most cases, the total release amount of radionuclides was usually not less than that of planned releases. Therefore, unplanned release is not negligible. The annual release amount listed in Table IV to Table VIII includes the contribution from unplanned releases.

2.4. Dose assessment

2.4.1. Dose calculation results

The main indices of dose assessment are individual effective dose equivalent to critical public group and collective effective dose equivalent to the population within a radius of 80 km. The distribution of the individual dose is given in Table X, in which the calculation unit is facility year. The distribution of the collective dose equivalent is shown in Table XI, the unit is the same in Table XI. Table XII is a list of annual average collective dose equivalent caused by the nuclear industrial systems.

2.4.2. Assessment of the calculation results

2.4.2.1. *Comparison with the national standards*

The assessment was made for the period from 1955 to 1985. During this period, the accordance with the national standard regulation for radiation protection was required. This standard was first issued in 1960 and then revised in 1974, in which the concept of critical group was not set forth clearly. There was only a similar concept of adjacent public, to whom the annual dose limit was 500 mrem (5mSv). Therefore, 5 mSv was used as an assessment criterion. The concept of critical group was proposed definitely in *Regulations for Radiation Protection* issued recently, and the annual dose limit is 1 mSv. This requirement was also taken into account in the assessment. It can be seen from Table X that the doses of the critical groups are lower than the dose limit for adjacent public (5mSv). It can also be seen from Table X that the doses for 93.5% of the facility years are lower than 1 mSv/a, and 77.1% lower than 0.25 mSv/a, 67.1% lower than 0.1 mSv/a. It can be concluded that the dose to adjacent public caused by all facilities is lower than the national dose limit, and the dose for most of the facilities is far below the limit. The facilities and the years for which the doses to the adjacent public exceed 1 mSv are enumerated in Table XIII. As Hengguang Uranium Mine in Hunan Province and Shangrao Uranium Mine in Jiangxi Province have been stopped, and the dose to critical group of Fuzhou Uranium Mine has dropped below 1 mSv/a since 1980, now all the nuclear facilities satisfy the requirement of the new standard.

TABLE X The distribution of annual individual effective dose equivalent (D , mSv) of critical group caused by each kind of nuclear facility

System	Distribution %			
	$D < 0.1$	$0.1 < D < 0.25$	$0.25 < D < 12.00$	$1.00 < D < 5.00$
Uranium mining & milling	31.3	8.06	14.8	6.2
Fuel fabrication	10.8			
Isotope separation plant	7.6			
Reactor & Reprocessing	5.8	1.0		
Research	11.6	1.0	1.6	0.2
Total	67.1	10.0	16.4	6.4

TABLE XI The distribution of collective dose equivalent (D , $man Sv$) caused by nuclear facilities

System	Distribution %								
	E-7 -E-6	E-6 -E-5	E-5 -E-4	E-4 -E-3	E-3 -E-2	E-2 -E-1	E-1 -E-00	E-00 -E-01	$D > 10$
Uranium Mining & Metallurgy					0.2	6.6	29.5	21.9	0.9
Fuel Fabrication			0.4		0.4	2.6	5.4	2.6	
Isotope Separation Plant	0.2	1.3	2.4	1.3	1.5	4.3	0.2		
Reactor & Reprocessing					0.7	2.4	3.0	0.9	
Research			1.7	1.9	1.7	1.7	2.2	1.9	0.2
Total	0.2	1.3	4.5	3.2	4.5	17.6	40.3	27.3	1.1

TABLE XII The distribution of annual average collective dose equivalent caused by the nuclear industrial systems

System		Collective Dose Equivalent	Distribution
Nuclear Fuel Cycle	Uranium Mining & Milling	1.93E1	83.3(91.5)
	Fuel Fabrication	1.09	4.7(5.2)
	Isotope Separation Plant	5.26E-2	0.2(0.3)
	Reactor & Reprocessing	6.47E-1	2.8(3.0)
	Total	2.10E1	91.00(100)
	Research & Isotope Production	1.93	8.4
	Nuclear Metallurgy & Processing	1.42E-1	0.6
	Total	2.31E1	100

TABLE XIII The facilities and the year of the dose of critical group in excess of 1 mSv

Facility	Pathway	Nuclide	Annual dose equivalent, Sv	Year
Hengyang Uranium Mine	inhalation	RN-222	1.01E-3	1983
	inhalation	RN-222	1.17E-3	1984
	inhalation	RN-222	1.31E-3	1985
Fuzhou Uranium Mine	inhalation	RN-222	1.16E-3	1977
	inhalation	RN-222	1.46E-3	1978
	inhalation	RN-222	1.20E-3	1979
Shangrao Uranium Mine	intake of crop	Pb210	1.12E-3	1975
	intake of crop	Pb210	1.27E-3	1976
	intake of crop	Pb210	1.30E-3	1977
	intake of crop	Pb210	1.35E-3	1978
	intake of crop	Pb210	1.40E-3	1979
	intake of crop	Pb210	1.44E-3	1980
	intake of crop	Pb210	1.50E-3	1981
	intake of crop	Pb210	1.63E-3	1982
	intake of crop	Pb210	1.74E-3	1983
	intake of crop	Pb210	1.78E-3	1984
	intake of crop	Pb210	1.82E-3	1985

TABLE XIV The distribution of relative collective dose equivalent of each kind of nuclear facility

System	Distribution %							
	1E-11 1E-10	1E-10 1E-09	1E-09 1E-08	1E-08 1E-07	1E-07 1E-06	1E-06 1E-05	1E-05 1E-04	1E-04 1E-03
Uranium Mining & Milling						2.6	29.2	16.7
Fuel Element Fabrication			0.2	0.2	0.7	3.1	5.6	2.8
Isotope Separation	0.5	1.2	2.6	1.4	1.7	4.7	0.2	
Reactor & Reprocessing						0.9	1.9	4.2
Metallurgy & Processing	1.0		1.7	2.4	1.9	0.2		
Research			1.7	2.8	2.1	2.8	1.7	1.4
Total	1.5	1.2	6.2	6.8	6.4	14.3	38.6	25.1

TABLE XV Annual collective effective dose equivalent due to some kinds of human activities (man Sv)

Human activity	Annual effective dose equivalent	Reference
Living in buildings built with stone coal cinder bricks	3.50E3	[1]
Living in buildings built with waste residue of aluminium processing plants	1.43E2	[2]
Living in buildings built in coal cinder from Xiangxi Gold Mine (only gamma radiation considered)	2.09E1	[3]
Travelling by civil flight	2.52E1	[4]
Whole nuclear industry	2.30E1	

2.4.2.2. *Comparison with the dose of national background radiation*

Early in the 1980's a nationwide survey of natural background radiation was started. According to the primary analysis of the survey results, the maximum difference for the annual dose of natural background radiation is about 1.10 mSv between the provinces and it would be greater between countries and much greater between persons. Thus it can be seen from Table X that the doses of the critical groups of the nuclear facilities are mainly within the fluctuation range of the average natural background.

Table XIV is a list of relative annual collective dose equivalent resulting from the nuclear facilities. The relative annual collective dose equivalent means that the percentage ratio of the collective dose equivalent resulting from a facility to the collective dose equivalent due to natural radiation was assumed to be 2.3 mSv. As shown in Table XIV, the relative collective dose equivalent of every nuclear system is below 0.1%, the relative collective dose equivalent of 85% of the nuclear facilities are within the range of $1.00E-6$ to $1.00E-3$, and the average public dose due to nuclear industry is about 0.001% of the average dose due to natural radiation in the area of interest.

2.4.2.3. *Comparison with collective effective dose equivalent resulting from other activities of human beings*

Annual effective dose equivalent of some kind of human activity is listed in Table XV. The annual collective dose equivalent to the public is $3.5E3$ man Sv as a result of living in buildings built with stone coal cinder bricks. That is about 150 times as much as that caused by the whole nuclear industry. The additional effective dose equivalent of gamma ray that was caused by using cement substitute made from waste residue of the three large aluminum plants in China is 6.2 times as much as that caused by the whole nuclear industry, since the radioactive material content in the red mud was quite high. The additional dose due to travelling by civil flights is a little higher than that due to the nuclear industry. There is hardly any difference between the dose resulting from the nuclear industry and that resulting from the building built in coal cinder bricks of Xiangxi Gold mine.

2.5. Measures to further improve the radiological environmental quality

2.5.1. Defining management object, carrying out total amount control, reinforcement of effluent monitoring, and making monitoring systems perfect

Although the dose of the critical group from any one of the facilities is below 1 mSv/a, it is still necessary to prevent the dose from exceeding 1 mSv/a. Further studies on the nuclear pathways should be carried out to draw out a conclusion as soon as possible. According to the principle of optimization of radiation protection, the possibility of reducing the dose to the critical groups should be further studied, and efforts should be made to keep the annual dose to the critical groups below 0.25 mSv in the near future.

Control of total amount of effluent may effectively protect the environment and is helpful to the development of production. Because the release amount is relatively high from uranium mining/milling plants and fuel fabrication factories, according to the principle of optimization of radiation protection, authorized limits of radioactive material released into the environment should be defined for each nuclear facility. In order to examine the consistence with the authorized limits, the regulations and rules about effluent monitoring should be established and equipment for effluent monitoring should be improved. Effluent monitoring is essential to the source term evaluation for environment assessment.

Environment monitoring of the nuclear industry has been carried out for over 30 years, but there are some evident shortcomings. For example, proper analysis methods are not available or have not been put into use in routine monitoring for the analysis of Pb-210, Po-210 and Ra-226 from uranium mining and milling system, and C-14 and airborne I-131 from such facilities as reactors.

TABLE XVI The annual average collective dose due to unplanned and planned releases of liquid effluent and their ratios

System	Time	Annual collective dose man Sv		Unplanned Planned
		Unplanned	Planned	
Fuel Element Fabrication	1973-1985	<u>3.69E-3</u>	<u>1.79E-3</u>	2.1
Isotope Separation	1962-1985	<u>2.18E-2</u>	<u>1.07E-2</u>	2.0
Reactor & Reprocessing	1967-1985	<u>4.96E-3</u>	<u>2.00E-4</u>	24.8
Research	1968-1986	7.06E-3	3.70E-4	19.1

2.5.2. Strengthening management and reducing release

It is clear from Table XVI that the dose caused by unplanned releases in every kind of nuclear facility except uranium mining and milling system (for which there is no detailed investigation data to be analysed). Therefore, it is a very important step to take effective measures to cut down accidental release for the lower total release amount. Lack of rigorous radioactive working area classification and shortage of strict rules about radioactive pollution monitoring are the important reasons for the high unplanned release amount.

The management of the existent waste treatment equipment should be improved. There are usually appropriate waste treatment equipment for gaseous and liquid effluent in all facilities, but quite a few of them are in abnormal operation status because of bad maintenance, and consequently the increase of release amount happened. It is because of the abnormal operation of filtration system that the collective effective dose caused by gaseous release from a plutonium metallurgical workshop in 1982 was higher than the dose caused by the total release within all the other years. The reinforcement of the management of waste treatment equipment is of significant importance for the decrease of the release amount.

2.5.3. Enhancing safety control to drop the frequency of accident

The collective dose resulting from accident releases is about 9% of the dose due to normal operation. This percentage might be higher if the dose caused by the loss of solid radioactive materials and radiation sources that were out of control. Practices prove that the frequency of accidents can be decreased as long as safety control is enhanced. The ratio of accident due to the dose of normal operation in every year is shown in Figure 1, from which it can be seen that dose decreases remarkably in the eighties as safety control became stricter. In order to improve safety control, it is necessary to carry out probability analysis and accident tree analysis and to perfect the record and report system of incidents and accidents.

2.5.4. Reinforcement of overall planning and optimization of investment benefit

The first thing is to analyse the risk in an all-round way and find main detriment factors for optimization of investment benefit.

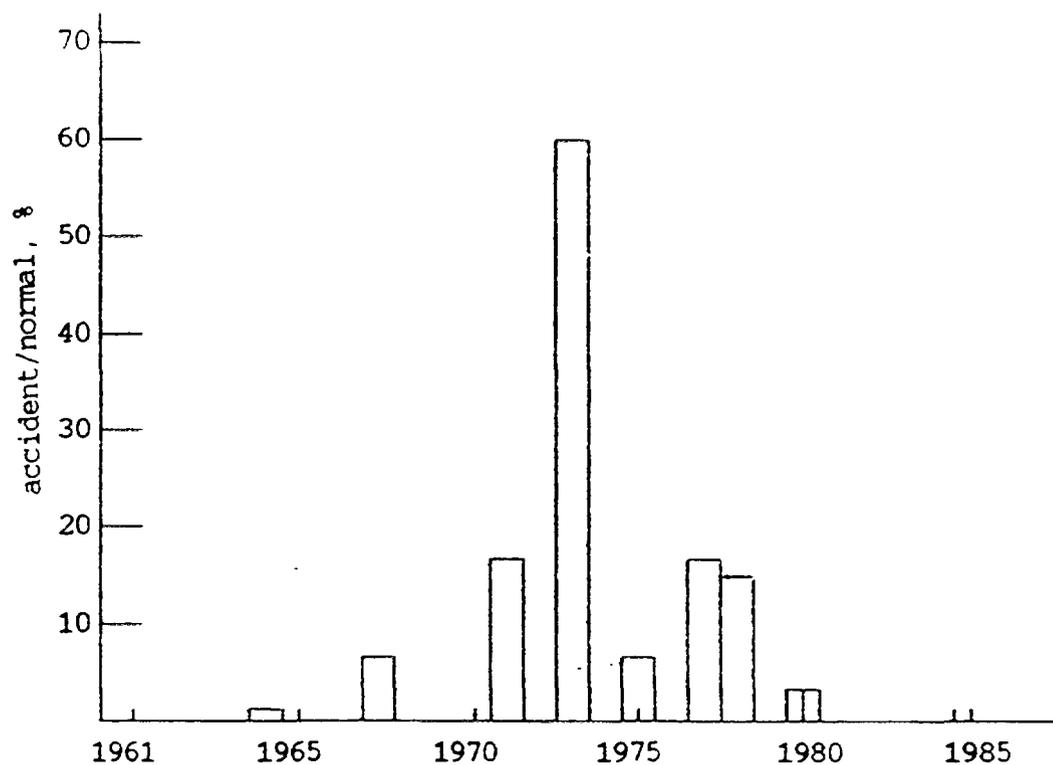


Figure 1. The ratio of accident of the dose of normal operation in every year

TABLE XVII The portions of gaseous effluent and liquid effluent in the annual average collective (man Sv) for each kind of nuclear facility

System	Gaseous		Liquid	
	Annual average	Portion %	Annual average	Portion %
Uranium Mining & Milling	1.47E01	76.6	4.95E00	23.4
Fuel Fabrication	1.09E00	99.6	4.00E-3	0.4
Isotope Separation	2.05E-2	39.0	3.21E-2	61.0
Reactor & Reprocessing	5.70E-1	88.1	7.68E-2	11.9
Research	3.50E-1	99.6	1.27E-3	0.4
Isotope Production	1.57E00	99.6	5.79E-3	0.4
Nuclear Metallurgy & Processing	1.41E-1	99.6	5.50E-4	0.4
Total	1.84E01	79.7	4.71E00	20.3

TABLE XVIII **The distribution of the annual average collective dose due to the nuclides released from the nuclear industry**

Nuclide	Gaseous		Liquid		Total	
	Dose man Sv	%	Dose man Sv	%	Dose man Sv	%
Natural U	1.99E-00	10.5	2.64E-01	5.7	2.25E-00	9.5
Enriched U	1.11-E01	0.5	2.90E-02	0.6	1.40E-01	0.6
Rn-222	1.39E+01	73.8	---	---	1.39E+01	59.0
Ra-226	3.01E-02	0.2	1.04E-00	22.3	1.07E-00	4.6
Th-230	2.64E-02	0.1	1.50E-02	0.3	4.14E-02	0.2
Po-210	1.47E-02	0.1	5.98E-02	1.3	7.45E-02	0.3
Pb-210	7.87E-02	0.4	3.22E-00	68.9	3.30E-00	14.0
Ar-41	1.41E-01	0.8	---	---	1.41E-01	0.6
Xe-133	3.08E-03	0.02	---	---	3.08E-03	0.01
Kr-85	6.70E-04	0.003	---	---	6.70E-04	0.002
I-131	1.52E-00	8.1	---	---	1.52E-00	6.4
I-125	3.00E-01	1.6	---	---	3.00E-01	1.3
Sr-90	3.21E-01	1.7	4.45E-02	0.9	3.66E-01	1.6
Pu-239	2.09E-01	1.1	7.02E-04	0.03	2.10E-01	0.9
Cs-127	1.57E-01	0.8	1.28E-03	0.1	1.58E-01	0.7
H-3	5.88E-02	0.3	2.32E-04	0.01	5.90E-02	0.3
Co-60	4.99E-03	0.02	4.74E-04	0.02	5.22E-03	0.03

TABLE XIX Nuclides listed in descent order according to their dose

	1	2	3	4	7	8	9
Total dose	Rn-222	Pb-210	Natural U	I-131	Ra-226	I-125	Sr-90
Gaseous	Rn-222	Natural U	I-131	I-125	Pu-239	Ar-41	Natural U
Liquid	Pb-210	Ra-226	Natural U	Sr-90	Po-210	Natural U	Th-230

The annual average collective doses from every kind of nuclear facility due to gaseous and liquid releases and the ratios between them are listed in Table XVII, the dose of gaseous effluent possesses 80.3% of the total dose i.e. 4 times as big as that of liquid effluent. In all the nuclear systems except uranium separation plants, the dose due to gaseous effluent is higher than the dose resulting from liquid effluent. Although the release of liquid radioactive materials caused higher dose than the gaseous release from uranium isotope separation plants, its total dose was not so high. Therefore, to study and tackle gaseous effluent is very important. In the past, more attention was paid to the tackling of liquid radioactive effluent, and less to gaseous effluent in many cases. In the uranium mining and milling system, for example, 34 000 000 Renminbi yuan was used to tackle liquid effluent, but only a little was used for gaseous effluent tackling.

The distribution of the annual average collective doses of each nuclide released from the nuclear industry is shown in Table XVIII. Table XIX is a list of the nuclides in a decent order of their dose according to Table XVIII. The distribution of the critical nuclides and critical pathways are given in Table XX. From the point of view of annual average collective dose and the dose of the critical group, the nuclides which need to be tackled are Rn-222, Pb-210, natural uranium, I-131, Ra-226, Pu-239, Sr-90, Cs-137, H-3, I-125 and Ar-41. Unfortunately, study on the Rn-222 and Pb-210 tackling has rarely been done.

2.6. Prediction of radiological environment quality in 2000

The radiological environment quality in 2000 may be predicted based on the development programme of the nuclear industry and with the consideration of the possible environmental protection measures to be taken in that period. The models used in the prediction are the same as those for the present situation assessment.

2.6.1. Prediction basis

- i) There will be 6.5 GW(e) nuclear power in the mainland, the spent fuel will be stored in spent fuel storage pools.
- ii) All the fuel elements needed by the nuclear power plants will be made in China and all the metallic uranium for fuel element manufacture will be produced in China.
- iii) Radioactive isotope production grows a lot, the output of radioactive iodine increases once.
- iv) Other nuclear facilities will be adjusted, some of them develop to some extent.

TABLE XX Distribution of critical nuclides and critical pathway

Nuclide	Rn-222	Pb-210			U				Pu-239	I-131	Ar-41	H-3
Pathway	Inhalation	Fish	Crop	Drink	Inhalation	Drink	Ingestion	Surface	Inhalation	Ingestion	immersion	Ingestion
Number of unit years	174	33	28	7	66	14	45	5	32	38	22	2
Portion %	37.3	7.1	6.0	1.5	14.2	3.0	9.7	1.0	6.9	8.2	4.7	0.4
		14.6			27.9							

TABLE XXI Cost-benefit analysis of environmental protection equipment and measures

Project	Cost 1.0E4 yuan	Benefit		$\Delta D/\Delta S$ 1.0E4 yuan/man Sv
		Net income, yuan	RCD man Sv	
Waste water treatment of Quzhou Mine	1.16		8.1 E-2	14.28
Waste water treatment of the 1st. mine in Southern China		8900	7.00E-3	
Waste water treatment of the 3rd. mine in Southern China		30100	0.42	
Waste water treatment of the 5th. mine in Southern China	16.6		4.55E-2	370
Relocation of local residents near the tailing dam of Hengyang Uranium Processing Mill	186		30	6.2
Air purification in processing systems of metallurgy	27.1		1.7	16.0
I-131 treatment in Institute of Atomic Energy	1.31		2.2	0.59
Installing 5 Ventura cleaners in a fuel fabrication plant	0.18		0.18	1.0
Treatment of low level uranium waste water in a fuel fabrication plant	4.98		3.6E-3	1400
Treatment of ammonium nitrate solution containing uranium		6309	2.8E-4	

2.6.2. The factors considered in prediction and the environmental protection measures to be possible taken

The factors considered in the prediction are:

- i) Annual increase rate of population is 0.6%.
- ii) Accidental dose will be 4.5% of the dose due to abnormal operation (except uranium mining and milling).

The following environment protection measures will be taken:

- i) In uranium mining and milling system, management will be improved, mined mine lanes will be blocked timely; 50% of tailing dams will be covered by a layer of soil (0.5 meter thickness); the concentration of Pb-210 and Ra-226 in waste water will decrease 50%.
- ii) In fuel element fabrication system, dust cleaning equipment will be improved, and the normalized amount of release will be cut down 50%.
- iii) In research and isotope production system, iodine filtration equipment will be installed for every radioactive iodine production installations; filtration efficiency will be up to 90%.

2.6.3. Prediction assessment results

The source term for the nuclear power plants in 2000 is anticipated upon the planned releases from Qinshan Nuclear Power Plant and Guangdong Nuclear Power Plant. The expected release amount from uranium mining and milling system, isotope separation installations and fuel fabrication factories, is based on the present release situation and the possible environment protection measures to be taken in the future. The results of the prediction of radiological environment impact in 2000 are given in Table XXII.

TABLE XXII Prediction of radiological environmental impact in the year 2000

System	Annual collective dose equivalent man.Sv
Uranium Mining & Milling	41.6
Fuel Fabrication	1.5
Isotope Separation	0.31
Nuclear power	11
Decommissioning tailing dam	1.4
Research & Isotope Production	1.9
Accident	1
Others	0.18
Total	58.9

2.7. Conclusions

The dose of the critical group of each nuclear facility is below the national limit and 93.6% are below 1 mSv/a. As a result of proper measures, there have been no facilities that cause doses higher than 1 mSv/a since 1986. The annual average dose due to natural radiation to the public in China is about 2.3 mSv. The dose of 80% of the critical groups near the nuclear facilities is lower than 1/10 of it, the highest dose of the critical groups is approximately 1/2 of it. In fact, the dose caused by every installation is within the fluctuation range of the average natural radiation dose of all the provinces of China.

The annual collective dose equivalent to the population in all the assessment areas is about 23 man.Sv. This is lower than 1/10000 of the natural radiation dose to the same population, and is lower than the collective dose caused by some non-nuclear facilities or other human activities. The annual collective dose resulting from living in buildings built in stone coal cinder bricks is two orders of magnitude higher than the collective dose caused by the whole nuclear industry. Compared with the detriments that naturally exist or are caused by other human activities, the detriment of the nuclear industry is negligible. The risk brought by the nuclear industry is 10 000 times lower than the risk by transportation, 100 000 lower than cancer death or 1000 lower than natural risk (for example, lightning strike).

Through the analysis of the present situation of nuclear industrial environment protection in the point of view of optimization of radiation protection, a lot of problems are found and need to be further solved. For instance, (1) compared with the data reported abroad, the amount of radioactive effluent released from some facilities, especially from uranium mining and fuel fabrication plants, is higher and unplanned releases is relatively high, too; in order to control or decrease the release amount, it is necessary to define an authorized limit of annual releases for each facility on the basis of this assessment, and to develop relevant tackling programme upon optimization analysis. (2) Although there is rather perfect environmental monitoring plan in each nuclear facility in China, quite a few of the vulnerable links and some spots that should be adjusted were found through this systematical assessment; they are mainly the selection of effluent monitoring method and quality control, the analysis of Pb-210, and Po-210 in waste water of uranium mining and milling facilities and the analysis of C-14 in the effluent from reactor facilities; on the basis of optimization, environmental monitoring programmes should be adjusted according to the purposes and requirement of environmental monitoring so as to improve monitoring quality. (3) The assessment of radioactive waste storage facilities was not done; the facilities might not impose significant effects on the environment under normal conditions, but they have tremendous potential risk, especially high and intermediate level waste liquid storage facilities. The potential environmental impact of radioactive material transportation was not taken into account. They need to be further studied and evaluated. (4) The reliability of the assessment models should be further studied and evaluated, the parameters should be systematically compiled and evaluated, and then a whole set of basic parameters that is appropriate for the specific situations in China will be achieved.

Based on the development programme of the nuclear industry in China and the possible measures to be taken, the collective dose to the people in all the assessment areas is predicted to be 60 man.Sv in 2000, which is 1.2/10 000 of the natural radiation dose to those people. The dose to critical groups will continue to decrease, and will be below 0.25 mSv in the year 2000. Therefore, compared with coal electricity, the acceleration of the development of nuclear power will be helpful to ameliorate the environment problems caused by the energy source development.

3. NON-RADIOLOGICAL ENVIRONMENTAL ASSESSMENT

The methods and models used for the non-radiological environmental assessment is similar to those for the assessment of radiological environment. In addition to contamination index method which is usually used in environmental assessment, the health hazard assessment method was used also for the purpose of an unified measure of the hazards from both radiation and non-radiation practices.

In addition to these harmful materials, such as fluorine and nitrogen oxides, exhausted from the main technological process, the contaminants exhausted from the power and heat supplying system firing

coal for the main technological process were also taken into account, including harmful materials SO₂, NO_x, F, B(a)P, Pb, Cd, As and Cr and radioactive materials U, Th, Ra, ²¹⁰Po and ²¹⁰Pb. The investigation on greenhouse gases was also made, such as CO₂, CH₄ and so on.

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RADIOACTIVE CONTAMINATION OF THE ENVIRONMENT IN THE AREAS OF LOCATION OF OBJECTS OF THE NUCLEAR FUEL CYCLE

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Abstract

The entry of radionuclides into the environment occurs at all stages of the Nuclear Fuel Cycle: during the extraction and reprocessing of uranium ore, production of fuel elements, operation of nuclear reactors, reprocessing of spent fuel, and burial of radioactive wastes. This report shows the generalization of data in the areas of location of objects of the nuclear fuel cycle (NFC) in the territory of Russia.

INTRODUCTION

Generalized data on the quantities and radionuclide composition of releases to the atmosphere from nuclear facilities of the Ministry of Atomic Energy (Minatom) of Russia are presented in Tables I and II [1, 2].

Among natural radionuclides, ^{222}Rn is a major contributor to the activity of atmospheric releases (about 420 TBq/year). Among artificial radionuclides, isotopes of inert radioactive gases (IRG) have the greatest activity in atmospheric releases of enterprises of Minatom (about 28 PBq). Releases of tritium amount to 410 TBq/year. An appreciable contribution to the IRG activity is made by gas-aerosol releases from NPPs. At the present time, 29 power units with a total installed capacity of 21.242 GWt (el.) are functioning in Russia. Table III presents normalized estimates of radioactive releases from NPPs in Russia in the period 1985-1993. In all, gas-aerosol releases from NPPs with the RBMK-type reactors are distinctly higher than those with the WWER-type reactors. There is a tendency for a decrease in the activity of atmospheric releases in the period 1991-1993, as compared to the preceding period 1985-1990, which is associated with the improvement of the gas purification system at NPPs.

Table IV presents the radionuclide composition of liquid discharges from enterprises of Minatom in 1992-1993 to the surface waters [1, 2]. In 1992, the total activity of liquid discharges amounted to 11.5 PBq. A major contribution to this activity was made by short-lived radionuclides (^{24}Na , ^{31}Si and ^{64}Cu) with a half-life less than 24 hrs, which accounted for about 81%, and by radionuclides with a half-life from 24 hrs to 1 month (^{32}P , ^{51}Cr , ^{56}Mn , ^{76}As , ^{239}Np and others), which accounted for about 18%. The activity of long-lived nuclides with a half-life over 1 year amounted to 9 TBq or less than 1% of the total activity of discharges. A major contribution to the activity of long-lived nuclides was made by tritium, ^{60}Co , ^{106}Ru , ^{90}Sr , ^{137}Cs and others. In 1993, a considerable decrease was observed in the activity of radioactive discharges to the surface waters, as compared to 1992. In 1993, the total activity of liquid wastes amounted to about 1.2 PBq, i.e. decreased by an order of magnitude, as compared to 1992. This decrease in the activity of radionuclide discharges was due to the reduced output at the enterprises of Minatom and removal from service of a number of nuclear facilities.

The bulk of radionuclides enters with liquid discharges from the enterprises of Minatom into the rivers of the Arctic basin (the Yenisei, Tom' and Techa Rivers) [1, 2].

TABLE I Releases of radionuclides into the atmosphere from the enterprises of Minatom in 1992-1993 [1,2], Bq/year

Composition of releases	1992 r.	1993 r.
Inert radioactive gases	3.0 E(16)	1.5 E(16)
Isotopes of iodine	1.6 E(11)	7.7 E(10)
Tritium	3.8 E(14)	3.9 E(14)
Isotopes of U, Pu and transuranic elements (natural and artificial)	8.5 E(10)	8.2 E(10)
Rn-222	3.6 E(14)	4.7 E(14)

Below are considered the characteristics of radioactive contamination of the environment during the operation of objects of NFC involved in the extraction of uranium ore and its enrichment, as well as the production of fuel elements, and reprocessing of irradiated nuclear fuel and radioactive wastes.

Argun' River Regional Mining and Chemical Production Association (MCPA)

This MCPA is located in Krasnokamensk district of Chita Region 400 km to the south-east of the city of Chita. It extracts uranium ore from open-cut and underground mines, and enriches it. The sources of radioactive contamination of the environment are the following: an open-cut pit raising dust, terraces of open-cut mines, a tail repository, and a hydrometallurgical plant on uranium ore enrichment. This MCPA also extracts coal and molybdenum ores, which results in chemical contamination of the environment [1, 2].

Mine waters from the Argun' River Regional MCPA are discharged to the surface storing reservoirs and water bodies of the open hydrographic system. Data on the amounts of radionuclides discharged in 1993 are given in Table V. Table VI presents some estimates of the radiation situation parameters in the zone of observations around the MCPA in 1993. The estimates presented indicate that the soil and vegetation radioactivity is mainly determined by natural radionuclide ^{40}K . The distribution of the total activity of beta emitters in water and atmospheric fallout samples within the area under observation is relatively uniform.

Production Association "Novosibirsk Plant of Chemical Concentrates" (PA "Chemconcentrate")

This PA is located in the northern part of the city of Novosibirsk [1]. It reprocessed uranium concentrates, and now it produces fuel elements for nuclear power engineering. A tail repository for burial of radioactive wastes containing radionuclides of the uranium series, lithium and mercury is located at a distance of 4 km from the PA. Radioactive wastes are delivered to the repository as sludge through the sludge pipeline. Before this pipeline was put into service, wastes were delivered by motor transport, and the environment was contaminated along the route of the delivery.

There are several potential sources of radioactive contamination of the environment at the PA "Chemconcentrate": atmospheric releases of natural and enriched uranium through ventilation systems; the entry of ^{222}Rn into the atmosphere from the surface layer of buried wastes; and the entry of radionuclides into the environment during accidents and the sludge pipeline failure.

TABLE II The radionuclide composition of the atmospheric releases from the enterprises of Minatom in 1991-1993 [1,2], Bq/year

Composition of releases	1991	1992	1993	Average
A. Natural radionuclides				
Enriched uranium	1.5 E(9)	1.2 E(9)	1.3 E(9)	1.3 E(9)
Natural uranium	1.0 E(9)	6.6 E(10)	6.1 E(10)	4.3 E(10)
U-238	2.8 E(10)	1.4 E(10)	1.4 E(10)	1.9 E(10)
Other alpha emitters	3.2 E(9)	7.8 E(8)	3.1 E(9)	2.4 E(9)
Rn-222	—	3.6 E(14)	4.7 E(14)	4.2 E(14)
B. Artificial radionuclides				
Na-24	4.4 E(8)	1.9 E(9)	—	1.2 E(9)
P-32	2.8 E(10)	5.2 E(10)	4.8 E(10)	4.3 E(10)
Sc-46	2.2 E(8)	1.9 E(8)	1.9 E(8)	2.0 E(8)
Cr-51	6.3 E(10)	6.8 E(10)	4.1 E(10)	5.7 E(10)
Mn-54, 56	9.6 E(8)	2.0 E(9)	1.9 E(9)	1.6 E(9)
Fe-55, 59	5.6 E(8)	5.6 E(8)	5.6 E(8)	5.6 E(8)
Co-57, 58, 60	2.3 E(9)	4.0 E(9)	4.1 E(9)	3.5 E(9)
Zn-65	1.2 E(9)	9.3 E(8)	8.9 E(8)	1.0 E(9)
Rb-88, 89	9.6 E(10)	7.6 E(10)	—	8.6 E(10)
Sr-89, 90	4.4 E(9)	5.7 E(9)	3.6 E(9)	4.6 E(9)
Zr-95 + Nb-95	2.6 E(10)	2.4 E(10)	1.6 E(10)	2.2 E(10)
Mo-99 + Tc-99	1.1 E(8)	3.7 E(9)	7.0 E(8)	1.5 E(9)
Ru-103, 106	2.3 E(10)	2.1 E(10)	2.3 E(10)	2.2 E(10)
Sb-124, 125	4.3 E(9)	1.1 E(8)	1.1 E(8)	1.5 E(9)
Cs-134, 137, 138	1.4 E(11)	1.1 E(11)	2.2 E(10)	9.1 E(10)
Ce-141, 144	1.7 E(10)	1.6 E(10)	1.2 E(10)	1.5 E(10)
Ba-139, 140	7.3 E(9)	4.7 E(9)	1.5 E(10)	9.0 E(9)
La-140	—	3.7 E(7)	—	3.7 E(7)
Bi-214	1.9 E(10)	3.1 E(9)	—	1.1 E(10)
Pb-212, 214	4.1 E(9)	2.2 E(8)	—	2.2 E(9)
H-3	4.5 E(14)	3.9 E(14)	3.9 E(14)	4.1 E(14)
Isotopes of I	1.2 E(11)	1.6 E(11)	7.7 E(10)	1.2 E(11)
Total activity of alpha emitters	4.3 E(9)	3.7 E(9)	3.0 E(9)	3.7 E(9)
Isotopes of Pu	3.7 E(8)	3.3 E(8)	3.3 E(8)	3.4 E(8)
Inert radioactive gases, including:	3.8 E(16)	3.0 E(16)	1.5 E(16)	2.8 E(16)
Ar-41	1.6 E(16)	1.3 E(16)	1.1 E(15)	1.0 E(16)
Long-lived nuclides	1.1 E(11)	1.1 E(11)	4.4 E(10)	8.8 E(10)
Short-lived nuclides	—	1.7 E(12)	2.8 E(11)	1.0 E(12)

TABLE III Radioactive releases from NPPs of Russia in the period 1985-1993 [1,5], GBq/GWt(el.)/year

Type of Reactor	Years	Inert radioactive gases	Long-lived nuclides	I-131
WWER	1985-1990	$(2.2 \pm 0.8) E(5)$	2.8 ± 0.2	1.3 ± 0.8
	1991-1993	$(5.6 \pm 4.0) E(4)$	1.7	1.0
RBMK	1985-1990	$(2.0 \pm 0.4) E(6)$	11 ± 7	8.0 ± 3.0
	1991-1993	$(1.7 \pm 0.9) E(5)$	4.6 ± 1.9	2.8 ± 1.4

Note: NPPs with the WWER type reactors are Balakov, Kalinin, Kola and Novovoronezh ; NPPs with the RBMK type reactors are Kursk, Leningrad and Smolensk.

Table VII presents some parameters for the assessment of the radiation situation in the zone of observations around the PA "Chemconcentrate" in 1992. According to observational data, the total activity of beta-emitting nuclides in the atmospheric fallout and the air was within the regional radiation background variations [1]. The exposure dose rate (EDR) in the 100-km zone of observations was 0.07-0.11 $\mu\text{Gy/hr}$ on the average, i.e. was also within the natural background variations. At the same time, anomalous patches were detected in the Kalinin district of the city of Novosibirsk, with levels of EDR as high as 2.1 $\mu\text{Gy/hr}$ around the PA "Chemconcentrate". A probable cause for the formation of these patches is the violation of regulations of solid radioactive waste transportation. Gamma radiation exposure dose rates in the area of location of the sludge pipeline were basically within the range from the background values to 0.35 $\mu\text{Gy/hr}$. A spectrometric analysis of soil samples collected in the area of location of the sludge pipeline indicated that they contained ^{226}Ra and ^{232}Th in amounts exceeding the background values. In the 5-km zone around the tail repository, the values of EDR varied from the background ones to 0.44 $\mu\text{Gy/hr}$. In the north-eastern part of the tail repository, a patch with an area of about 3 m^2 and a gamma radiation level approximately 9 $\mu\text{Gy/hr}$ was detected.

The activities of beta emitters in potable ground waters are characterized by high variability and vary from 11 to 440 Bq/l. At the present time, it is difficult to make any unambiguous inference about contamination of the ground waters around the tail repository for lack of the system of radioactive contamination monitoring of ground waters.

Siberian Chemical Industrial Complex (SCIC)

This industrial complex is located in the town of Tomsk-7 on the right bank of the Tom' River 15 km to the north of the city of Tomsk. It is the largest complex on the production of plutonium, uranium and transuranic elements. The SCIC comprises the following plants which are the sources of radioactive and chemical contamination of the environment:

- a reactor plant with commercial uranium-graphite reactors for the production of weapon-grade plutonium and electric energy;
- a plant on isotope separation for the production of enriched uranium hexafluoride;
- a plant on production of uranium protoxide-monoxide and uranium hexafluoride;
- a radiochemical plant on reprocessing of irradiated standard lumps to obtain and treat uranium and plutonium salts;
- chemical-metallurgical works.

TABLE IV Discharges of radionuclides into the surface waters from the enterprises of Minatom in 1992-1993, Bq/year

Radionuclide	1992	1993
H-3	4.0 E(12)	3.3 E(12)
Na-24	7.5 E(15)	9.5 E(14)
Si-31	1.6 E(15)	2.4 E(13)
P-32	2.2 E(14)	4.6 E(13)
Sc-46	1.7 E(12)	1.3 E(11)
Cr-51	1.3 E(14)	8.3 E(12)
Mn-54	1.3 E(12)	1.7 E(10)
Mn-56	1.5 E(15)	9.0 E(13)
Fe-59	3.2 E(12)	5.1 E(10)
Co-58	9.4 E(12)	7.9 E(10)
Co-60	2.9 E(12)	1.1 E(11)
Cu-64	2.2 E(14)	1.1 E(13)
Zn-65	2.4 E(12)	7.0 E(10)
As-76	1.2 E(14)	3.0 E(12)
Sr-90	6.1 E(11)	1.1 E(12)
Zr-95	5.0 E(11)	4.9 E(10)
Nb-95	3.4 E(11)	5.2 E(10)
Ru-103	3.4 E(11)	1.0 E(10)
Ru-106	1.3 E(12)	2.6 E(10)
I-131	2.2 E(12)	6.1 E(10)
Cs-134	1.9 E(9)	2.0 E(9)
Cs-137	2.2 E(11)	6.3 E(10)
Ba-140	1.7 E(12)	5.2 E(10)
Ce-141	6.1 E(11)	1.7 E(10)
Ce-144	7.1 E(11)	1.0 E(11)
Pb-210	3.0 E(9)	2.1 E(9)
Po-210	1.2 E(10)	1.1 E(10)
Ra-226	5.4 E(9)	4.1 E(9)
Np-239	1.7 E(14)	8.7 E(12)
Pu-239	1.6 E(8)	--
Total activity including:	1.154 E(16)	1.16 E(15)
a) Short-lived nuclides (T ½ < 24 hrs)	9.4 E(15)	9.85 E(14)
b) Radionuclides with 24hrs < T ½ < 30 days	2.1 E(15)	1.7 E(14)
c) Long-lived nuclides (T ½ > 1 month)	3.0 E(13)	5.2 E(12)
d) Long-lived nuclides (T ½ > 1 year)	9.0 E(12)	4.6 E(12)

TABLE V Discharges of radionuclides with sewage waters from the Argun' River Regional MCPA in 1993, Bq/year

Radionuclide	Total	Natural water bodies	Technological storage ponds
²⁴⁰ Pb	1.6 E(10)	2.1 E(9)	1.4 E(10)
²¹⁰ Po	8.0 E(9)	1.6 E(9)	6.4 E(9)
²²⁶ Ra	1.8 E(10)	2.7 E(9)	1.5 E(10)
Th	7.4 E(9)	2.5 E(9)	4.9 E(9)
U	9.9 E(9)	3.5 E(9)	9.6 E(11)

TABLE VI Parameters for the radiation situation assessment in the zone of observations around the Argun' River Regional MCPA in 1993

Parameter (object) of assessment	Dimension	Average value	Maximum value
1. Exposure dose rate	$\mu\text{Gy/hr}$	$0.11 \pm 0.02(14)$	0.14
2. Specific activity of radionuclides			
Soil	Bq/kg		
⁴⁰ K		$820 \pm 190 (14)$	1100
¹³⁷ Cs		$40 \pm 26 (6)$	80
²²⁶ Ra		$60 \pm 9 (14)$	78
²³² Th		$29 \pm 4 (11)$	35
Vegetation	Bq/kg		
⁴⁰ K		$950 \pm 510 (11)$	1900
Water			
Total activity of beta emitters	Bq/l	$1.6 \pm 1.2 (14)$	4.4
Atmospheric fallout (total activity of beta emitters)	Bq/m ² per day	$3.0 \pm 0.9 (60)$	7.2

Note: The number of measurements is indicated in paranthesis. A radius of the zone of observations on the activity of the atmospheric fallout is 100 km and on the other parameters 20 km.

TABLE VII Some parameters for the radiation situation assessment in the zone of observations around the Novosibirsk PA "Chemconcentrate" in 1992

Parameter of assessment	Dimension	Average value	Maximum value
Exposure dose rate	$\mu\text{Gy/hr}$	0.09 ± 0.02 (11000)	2.1
Average monthly activity of beta emitters in the atmospheric fallout	Bq/m^2 per day	1.0 ± 0.7 (24)	9.4
Average monthly activity of beta emitters in the atmospheric air	$\mu\text{Bq/m}^3$	90 ± 23 (12)	180
Total activity of beta emitters in ground waters	Bq/l	100 ± 90 (9)	440

Note: The number of measurements is indicated in parenthesis. A radius of the area of observations on the exposure dose rate is 100 km and on the activity of ground waters 30 km

TABLE VIII Releases and discharges of radionuclides from the Siberian Chemical Industrial Complex in 1992 [1,7] Bq/year

Composition of releases (discharges)	Activity
Releases into the atmosphere	
Inert radioactive gases	7.6 E(15)
^{90}Sr	2.3 E(9)
^{131}I	1.6 E(10)
Total activity of alpha emitters ($^{234}, ^{235}, ^{238}\text{U}, ^{239}\text{P}, ^{241}\text{Am}$)	3.0 E(9)
Discharges to the Tom' River	
^{32}P	4.7 E(13)
^{24}Na	7.1 E(14)
^{46}Sc	1.0 E(11)
^{51}Cr	2.0 E(12)
^{60}Co	1.8 E(10)
^{239}Np	5.8 E(12)

The commercial channel-type reactors with graphite moderators were commissioned in 1958-1963. Three of them were shut down in 1990-1992, and the remaining two should operate till 1995. Since the start-up of the reactors, weapons-grade plutonium was produced, and fuel elements and spent nuclear fuel were reprocessed [6, 7].

Table VIII presents the quantities of radioactive releases and discharges from the SCIC in 1992. As a rule, the atmospheric releases were only fractions of a percent of the maximum permissible releases (MPR). However, for the water discharges of ecologically significant nuclides, such as ^{24}Na and ^{32}P , the discharged activity values were close to the maximum permissible discharges (MPD) or slightly exceeded them. An excess over MPD for ^{32}P in 1992 was mainly due to the lack of installations for desalinization of water supplied for nuclear reactor cooling. Putting into operation of such an installation in 1993 made it possible to decrease appreciably the activity of the water discharges of ^{32}P .

TABLE IX Parameters for the radiation situation assessment in the zone of observations around the Siberian Chemical Industrial complex in 1992

Parameter of assessment	Dimension	Average value	Maximum value
Exposure dose rate	$\mu\text{Gy/hr}$	0.10 ± 0.01	0.12
Average monthly activity of beta emitters in the atmospheric fallout	Bq/m^2 per day	0.74 ± 0.28 (109)	1.8
Soil (in the Tom' River floodplain)	Bq/kg		
⁵¹ Cr		80 ± 60 (4)	465
⁵⁴ Mn		18 ± 9 (13)	65
⁵⁹ Fe		12 ± 5 (2)	78
⁶⁰ Co		53 ± 26 (12)	265
⁶⁵ Zn		190 ± 100 (12)	950
¹³⁷ Cs		43 ± 30 (18)	640
¹⁴¹ Ce		14 ± 6 (7)	23
¹⁴⁴ Ce		54 ± 26 (2)	80
¹⁵² Eu		9 (1)	—
Total beta emitters		400 ± 260 (23)	1460
Exposure dose rate	$\mu\text{Gy/hr}$	0.16 ± 0.10 (24)	0.75
Grass	Bq/kg		
⁵⁴ Mn		68 (1)	—
⁶⁰ Co		39 (1)	—
⁶⁵ Zn		680 (1)	—
¹³⁷ Cs		100 ± 60 (2)	160
Needles	Bq/kg		
⁵⁴ Mn		120 (1)	—
⁶⁵ Zn		640 ± 360 (3)	890
¹³⁷ Cs		76 (1)	—
Total beta emitters in the Tom' River water	Bq/l	2.0 ± 1.2 (23)	3300
Ground Waters (wells of the Kantes plot)	Bq.m^3		
⁹⁰ Sr		36 ± 5 (2)	41
		22 ± 7 (2)	29

Note: The number of measurements is indicated in parenthesis.

The SCIC is also the source of chemical contamination of the environment. In 1992, discharges of hazardous chemicals (in % of MPD) amounted to 2.4 for fluorides, 1.7 for ammonia, 0.5 for nitrogen oxides, 0.2 for nitric acid, 0.016 for tributyl phosphate, 0.008 for paraffins, 0.001 for carbon tetrachloride, 0.008 for acetone, and 0.001 for benzene [7].

Fifty repositories of liquid and solid radioactive wastes (RW) are located in the territory of the SCIC. In open storage facilities, 4.7 E(18) Bq of RW have been accumulated, and in underground strata, 1.5 E(19) Bq of liquid RW have been pumped at a depth up to 300 m [7, 8].

Table IX presents some parameters of the radiation situation in the zone of observations around the SCIC in 1992. The values of EDR activity of beta emitters in the atmospheric fallout in the 100-km zone around the SCIC do not differ significantly from the regional radiation background. At the same time, radioactive contamination of water and soil is observed in some parts of the floodplain of the Romashka and Tom' Rivers where radioactive discharges from the SCIC have entered for a long time. The presence of ^{51}Cr , ^{54}Mn , ^{59}Fe , ^{60}Co , ^{65}Zn , ^{141}Ce , ^{144}Ce and ^{152}Eu was detected in soil samples collected in the Tom' River floodplain. Technogenic radionuclides ^{54}Mn , ^{60}Co and ^{65}Zn which are typical of discharges from the SCIC were detected in samples of vegetation (grass and needles). Technogenic radionuclides were also found in ground waters in amounts no greater than the permissible levels.

On April 6, 1993 at 12.58 local time, an accident with the release of radionuclides into the environment occurred at the radiochemical plant of the SCIC.

The total area of contamination with levels of EDR higher than a radiation background of 0.09 $\mu\text{Gy/hr}$ (10 $\mu\text{r/hr}$) was estimated in April 1993 at about 100 km^2 [2]. The dominant radionuclides in snow samples from the contaminated area were ^{95}Zr , ^{95}Nb , ^{106}Ru and ^{103}Ru . The traces of ^{239}Pu and ^{144}Ce were detected as well. A nonuniform structure of the field of radioactive contamination of the area determined by the presence of hot particles in the composition of radioactive products of the accident deposited onto the snow was revealed [2]. According to observational data, radioactive contamination of the environment with the products of the accident at the SCIC was of a local nature. The accident resulted in the formation of a narrow radioactive trace 35-45 km long in a north-eastern direction from the SCIC (from the trace concentrations of ^{95}Zr and ^{95}Nb in soil). There are no populated places in the territory of the trace, except for the village of Georgievka. The external exposure dose to inhabitants of Georgievka from the products of the accident over 50 years of permanent residence will amount to 0.22-0.31 mSv which is negligible, as compared to the dose from the natural radiation background. On a whole, the radiation accident at the SCIC has not led to significant radiological consequences for the population [2, 7].

In the process of gamma survey of the area around the SCIC, sites contaminated with ^{137}Cs up to 37-74 kBq/m^2 were detected. They fan out within 10 km of the SCIC to the north, 2-3 km to the west and east, as well as along the shore line of the Tom' River. The nature of this contamination does not allow relating it to the accident occurred in April 1993. Most likely, this contamination is the result of the SCIC operation of many years [2].

In conclusion, it should be pointed out that it is essential to develop an integrated radioecological monitoring of the radionuclide content in the atmosphere, soil, surface and ground waters, natural and agricultural food chains. Data of the radioecological monitoring are necessary for further analysis of risks posed by radioactive and chemical contamination of the environment, and for the development of immediate measures to insure ecological safety during the operation of enterprises of NFC in Russia.

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