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THE CHERNOBYL REACTOR ACCIDENT SOURCE TERM

DEVELOPMENT OF A CONSENSUS VIEW

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THE CHERNOBYL REACTOR ACCIDENT SOURCE TERM

Development of a Consensus View

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

Paris 1996

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ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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- *assessing the contribution of nuclear power to the overall energy supply by keeping under review the technical and economic aspects of nuclear power growth and forecasting demand and supply for the different phases of the nuclear fuel cycle;*
- *developing exchanges of scientific and technical information particularly through participation in common services;*
- *setting up international research and development programmes and joint undertakings.*

In these and related tasks, NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has concluded a Co-operation Agreement, as well as with other international organisations in the nuclear field.

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COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

The Committee on the Safety of Nuclear Installations (CSNI) of the OECD Nuclear Energy Agency (NEA) is an international committee made up of senior scientists and engineers. It was set up in 1973 to develop, and co-ordinate the activities of the Nuclear Energy Agency concerning the technical aspects of the design, construction and operation of nuclear installations insofar as they affect the safety of such installations. The Committee's purpose is to foster international co-operation in nuclear safety among the OECD Member countries.

The CSNI constitutes a forum for the exchange of technical information and for collaboration between organisations which can contribute, from their respective backgrounds in research, development, engineering or regulation, to these activities and to the definition of the programme of work. It also reviews the state of knowledge on selected topics on nuclear safety technology and safety assessment, including operating experience. It initiates and conducts programmes identified by these reviews and assessments in order to overcome discrepancies, develop improvements and reach international consensus on technical issues of common interest. It promotes the co-ordination of work in different Member countries including the establishment of co-operative research projects and assists in the feedback of the results to participating organisations. Full use is also made of traditional methods of co-operation, such as information exchanges, establishment of working groups, and organisation of conferences and specialist meetings.

The greater part of the CSNI's current programme is concerned with the technology of water reactors. The principal areas covered are operating experience and the human factor, reactor coolant system behaviour, various aspects of reactor component integrity, the phenomenology of radioactive releases in reactor accidents and their confinement, containment performance, risk assessment, and severe accidents. The Committee also studies the safety of the nuclear fuel cycle, conducts periodic surveys of the reactor safety research programmes and operates an international mechanism for exchanging reports on safety related nuclear power plant accidents.

In implementing its programme, the CSNI establishes co-operative mechanisms with NEA's Committee on Nuclear Regulatory Activities (CNRA), responsible for the activities of the Agency concerning the regulation, licensing and inspection of nuclear installations with regard to safety. It also cooperates with NEA's Committee on Radiation Protection and Public Health and NEA's Radioactive Waste Management Committee on matters of common interest.

The opinions expressed and the arguments employed in this document are the responsibility of the authors and do not necessarily represent those of the OECD.

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FOREWORD

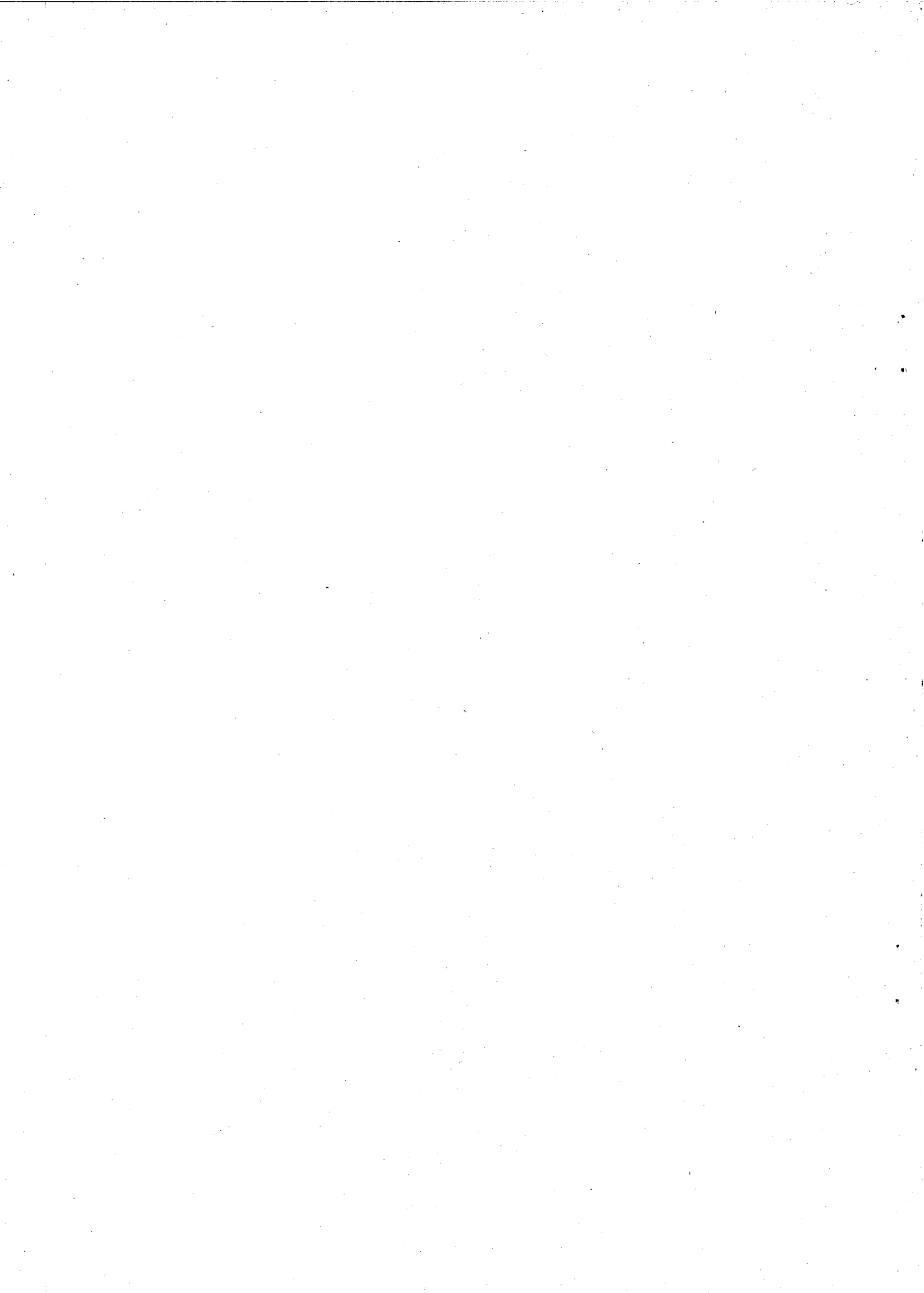
1996 will mark the tenth anniversary of the Chernobyl accident. The NEA Committee on Radiation Protection and Public Health (CRPPH) is preparing for that occasion an authoritative collective statement on the situation at Chernobyl, reviewing the current state of knowledge regarding the short-term and long-term health impact of the accident, discussing remaining problems (e.g., land contamination, population exposure), and presenting the lessons learned by the OECD Member countries in terms of radiation protection infrastructure and emergency preparedness. In order to make a full assessment of health effects, the CRPPH needed to consider dosimetric assessment. This implied, possibly, a re-evaluation of the magnitude of the Chernobyl accidental release. The CRPPH asked the Committee on the Safety of Nuclear Installations (CSNI) and the Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG-4) to discuss this matter and come up, if necessary, with revised estimates for the release.

Mr. L. Devell (Studsvik Eco & Safety AB, supported by SKI, Sweden), Mr. S. Guntay (Paul Scherrer Institut, Switzerland) and Dr. D.A. Powers (Sandia National Laboratories, USA) took the lead on the preparation of the following report, with ad hoc support from PWG-4's Task Group on Fission Product Phenomena in the Containment (FPC).

The report summarizes the recent general findings and consensus concerning the Chernobyl source term.

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EXECUTIVE SUMMARY

BACKGROUND

As a request from NEA/CRPPH to expert groups within CSNI the present report on the source term was prepared to be used as one input for a CRPPH report on Chernobyl radiological and health impact.

At the IAEA Post-Accident Review Meeting in Vienna 25-29 August 1986, the former USSR State Committee on the Utilization of Atomic Energy presented initial information on the source term. The source term is a technical expression for the release of radioactive material to the environment outside the reactor building. Important ingredients of the source term are amount of radioactive materials released (radionuclides and radioactivity) their physical and chemical forms and distribution over time.

QUANTITATIVE ESTIMATES OF RELEASES TO THE ENVIRONMENT

The initial information of the quantitative estimate of the release was mainly based on the integration of ground deposits within the territories of the former Soviet Union. It was made clear at the meeting in Vienna by USSR representatives that the estimate was based on this integration. In the discussions it was accordingly suggested that the total release of radiologically important nuclides therefore must be significantly higher if one considers also the deposition outside the former Soviet Union. Later, closer examinations of world wide distribution of e.g. cesium support that statement.

For the noble gases (xenon and krypton) the release was initially assumed to be 100 % of the core inventory and there are no results or discussion objecting to this assumption. For the more volatile elements iodine, tellurium and cesium the initial estimates varied between 10 and 20 %. For less volatile elements the estimates were between 3 and 6 %. For the fuel material dispersed outside the reactor building the early estimate was 3 ± 1.5 %.

The IAEA International Nuclear Safety Advisory Group issued in September 1986 its Summary Report INSAG-1 on the Post-Accident Review Meeting on the Chernobyl Accident. Release figures were based on those given in the USSR report to IAEA.

In the 1988 UNSCEAR report (United Nations Scientific Committee on the Effects of Atomic Radiation), the release figures are given according to the USSR report based on analyses of samples within USSR but also according to an estimate based on deposition all over the globe. The total release for Cs 137 was thus estimated to 70 PBq ($1 \text{ PBq} = 10^{15} \text{ Bq}$) compared to $39 \pm 19 \text{ PBq}$ in the USSR report based on integration within the borders of the former Soviet Union.

Russian analyses of core debris and deposited material inside the reactor building have made it possible to carry out an independent estimate of the release of cesium to the environment. From this type of analyses, Cs 137 release fraction to the environment was estimated by Russian scientists to be 33 ± 10 % ($2.3 \pm 0.7 \text{ MCi} = 85 \pm 26 \text{ PBq}$) based on an average release fraction from fuel of 47 % and a subsequent retention in the reactor building.

For the I 131 release, Russian scientists present estimates between 20 and 80 % of core inventory. The most accurate estimate is believed to be 50 to 60 % of the core inventory which corresponds

to a total activity of 1.1 to 1.3 EBq (1 EBq = 10^{18} Bq) of I 131 released into the atmosphere. The current best estimate for release of fuel from the reactor in the form of fragments is 3.5 ± 0.5 %.

RELEASE INTENSITY PATTERN

The initial USSR report gives an estimate of the release intensity pattern during the period from April 26 to May 6. After May 6, only very small releases of radioactivity are thought to have occurred. There is not much further information published concerning the release pattern and the measurements it is based upon. This is one reason of uncertainty concerning this matter. Another is the obvious difficulties of obtaining representative air samples. However, it is generally agreed that a substantial release occurred during the violent phase of the first day. Thereafter, the release rate decreased. Release began to increase from May 1 to May 6. A sudden arrest in the release of radioactivity after May 6 may have been due to a sharp reduction in fuel temperature as core debris interacted with structures in the lower regions of the reactor.

CHEMICAL AND PHYSICAL FORMS

The release of radioactive material to the atmosphere from the reactor occurred in the form of gases, aerosols and finely fragmented fuel. Volatile elements evaporated more or less from the fuel material due to the temperature increase during the accident. Krypton and xenon, which are noble gases, were considered to be completely released. Iodine appeared in the plume both in gaseous form and in particulate form. Organically bound iodine was also observed. The ratios between the various iodine forms varied in time. Other volatile elements, e.g., cesium and tellurium (and their compounds) condensed and were transported as aerosols independent of fuel material particles. A review of air sampling results in distant countries revealed that the particles containing iodine, cesium and tellurium had equivalent diameters in the range of 0.5-1 μ m.

Unexpected features of the Chernobyl source term were the extensive releases of fuel material and the extended release duration. Low volatile elements, e.g., cerium, zirconium, the actinides and to the major part also barium, lanthanum and strontium were embedded in fuel particles. Larger fuel particles deposited in the immediate neighbourhood. Smaller fuel particles were dispersed more widely.

A special peculiarity was the occurrence of ruthenium particles in the fallout. In addition there were observations of increased release of ruthenium and molybdenum relative to that of fuel material in the late phase of the emission period. These observations are thought to be due to formation of volatile oxides of these elements under the influence of air and subsequent transformation to more stable forms.

GENERAL REMARK

A number of more recent publications and results from scientists in Russia and elsewhere have significantly improved our understanding of the Chernobyl source term since 1986. Because of the special features of the RBMK reactor design and the accident peculiarities this understanding has however very limited application on other reactor types.

I INTRODUCTION

The worst accident ever experienced at a nuclear electric generating station occurred April 26, 1986 at Unit 4 of the Chernobyl Atomic Energy Station. This accident captured the attention of the world because radioactivity released from the damaged plant was detected in most countries in the northern hemisphere. In August 1986, scientists from the former Soviet Union provided the nuclear safety community with an impressively detailed account of what was then known about the Chernobyl accident [1]. This included assessments of the magnitudes, rates, and compositions of radionuclide releases during the ten days following initiation of the accident. A summary report based on the Soviet report, the oral presentations, and the discussions with scientists from various countries was issued by the International Atomic Energy Agency [2] shortly thereafter.

The account presented by the Soviet scientists did much to clarify the causes and the acute consequences of the Chernobyl accident. It was, however, a preliminary report prepared only shortly after the accident. Recovery actions at the station were still underway. Many topics of interest could not be explored at the time because affected parts of the nuclear reactor were still inaccessible.

Almost ten years have elapsed since the reactor accident at Chernobyl. A great deal more data is available concerning the events, phenomena, and processes that took place. The purpose of this document is to examine what is known about the radioactive materials released during the accident. This is a task that is substantially more difficult than it might first appear to be. The Chernobyl atomic energy station, like other nuclear power plants, was not instrumented to characterize a disastrous accident. The accident was peculiar in the sense that radioactive materials were released, at least initially, in an exceptionally energetic plume and were transported far from the reactor site. Release of radioactivity from the plant continued for several days. Characterization of the contamination caused by the releases of radioactivity has had a much lower priority than remediation of the contamination. Consequently, an assessment of the Chernobyl accident source term must rely to a significant extent on inferential evidence.

The assessment presented here begins with an examination of the core inventories of radioactive materials. In subsequent sections of this report, the magnitude and timing of the releases of radioactivity are described. Then, the composition, chemical forms, and physical forms of the releases are discussed. A number of more recent publications and results from scientists in Russia and elsewhere have significantly improved our understanding of the Chernobyl source term. Because of the special features of the reactor design and the peculiarities of the Chernobyl accident, the source term for the Chernobyl accident is of limited applicability of the safety analysis of other types of reactors.

II CORE INVENTORIES

A first step in the definition of a reactor accident source term is the determination of the inventory of radioactive materials that could be released from a nuclear plant. There have been a number of attempts to estimate the radionuclide inventories of the Chernobyl reactor at the time of the accident. Early attempts were handicapped by imprecise knowledge of the fuel burnup in the reactor. Begichev et al. [3] have clarified the burnup history of fuel in the reactor. Their estimates of burnup are shown in Table 1. Sich [4] has assembled data on samples of core debris remaining in the reactor and concludes that on average the burnup is about 11660 ± 650 MWd/t.

Various estimates of the radionuclide inventories are listed in Table 2. Inventories attributed in this table to Warman [5] were derived from data provided by INSAG [2] which were based on the

Soviet report [1]. These estimates have been critically reviewed by Clough [7] and by Devell [8], who noted that typographical errors may be responsible for overestimation of the ^{99}Mo inventory by a factor of 10 and underestimation of the ^{239}Np inventory by a factor of 10. Corrected values are shown parenthetically in the table. Clough provided comparison inventories obtained with the FISPIN code assuming average fuel burnups of 10300 and 13500 MWd/t. He also cited results obtained with the ORIGEN2 code by Anttila [6], who assumed an average burnup of 10000 MWd/t for fuel in seven burnup ranges from 2500 to 17500 MWd/t. Kichner and Noack [9] used the ORIGEN code to obtain inventories for fuel with an average burnup of 12850 MWd/t. Little [10] used the ORIGEN code and an average burnup of 10300 MWd/t to estimate core inventories. Begichev [3] used the varying fuel burnups listed in Table 1 to obtain estimates of the core inventories of radioactive materials. These burnup histories and the WIMS/CACH2 codes were used by Sich [4] to estimate inventories. The ^{136}Cs inventory reported by Sich was found to be in error and a corrected value [11] is shown parenthetically in Table 2.

There is general agreement among the various estimates of core inventories shown in Table 2. There are, however, large-enough variations that some caution is needed when inventories are used to calculate release fractions. Estimates of inventories made by Begichev et al. [3] may be the most reliable since they were calculated with the most detailed fuel history. Notable differences between results obtained by Begichev et al. and others are that the ^{132}Te and ^{89}Sr inventories are 30 to 40 percent lower and the ^{106}Ru inventory is 40 to 50 percent higher.

III RELEASE CHARACTERISTICS

The pattern of radioactivity release during the Chernobyl accident has been discussed by many authors [1-5, 12]. There was an initial, intense, release of radioactivity during the dynamic events following the reactivity insertion that started core disruption. This initial release included fragments of fuel as well as other types of aerosol particles, radionuclide vapors and noble gases. Though the released, radioactive material was lofted high above the reactor in an energetic plume, much of the material and especially the fragmented fuel particles, deposited within the borders of the former Soviet Union. Nevertheless, sufficient radioactive material, including material composed of fuel fragments, was carried beyond these borders to trigger what became a world-wide reaction to the accident.

The initial, intense phase of the accident was not the end of radioactivity releases from Chernobyl. Releases of radionuclides continued for several days after the initial core disruption. The rates of radioactive material release declined rapidly after the initial dynamic stage of the accident. A broad minimum in the radioactive material release rate was reached about April 29. Then, radioactive material releases began to increase despite heroic efforts at the reactor site to mitigate and manage the accident. On or about May 5, some 9-10 days after accident initiation, the radioactive material release rates dropped by about 3 orders of magnitude. Release rates that had been on the order of 10^{16} Bq per day fell to an average estimated by Cambrey et al. [23] to be 9×10^{11} Bq per day, though there may have been episodic eruptions of a few thousand curies per day over the next few weeks [1, 4]. Buzulukov and Dobrynin indicate significant releases of radioactivity occurred 20-21 and 25-30 days after accident initiation [13]. These releases may have been the result of vaporization of dust-suppression solutions admitted to the reactor vault. The releases are thought not to have added greatly to the total radioactivity release from the plant.

Quantitative descriptions of the radioactivity releases during the accident were presented by Soviet scientists in 1986 based on air sampling and surveys of ground contamination within the Soviet Union. It is known that estimates of the release are based mainly on surveys of ground

contamination [1, 4]. Materials that went outside the borders of the Soviet Union were not included. Noble gas releases were assumed to be complete.

It is not possible to confirm the results of early ground contamination surveys because of continued recontamination as the accident progressed. It is possible to confirm integral releases by comparison to radionuclide inventories in the reactor core debris. This has been done in the case of cesium as discussed later in this paper. In principle, it might be possible to confirm some aspects of the quantitative estimates of the release by analyzing radioactivity detected outside the Soviet Union. Such analyses would be quite difficult because of the shifting winds at the reactor site and rainfall along the transport path of the plume. Without more detailed information on the quantitative estimates of day-to-day releases of radioactivity during the Chernobyl accident, it is probably better to rely on the qualitative release pattern and the integral release estimates.

It has been assumed that the variations in the rates of radioactivity release could be explained by the behaviour of core debris within the reactor. It has long been known that a fairly vigorous natural circulation of air up through the damaged portion of the reactor provided an efficient transport of radionuclides released from the core debris. Further interpretation of the release has been complicated by uncertainties concerning the behaviour of the graphite moderator and the effects of many tons of lead, borax, clay, and sand dropped into the reactor vault to smother the burning graphite.

In recent years, it has been possible to re-enter the reactor, take samples, make measurements, and clarify some of the effects of deposited materials and to determine the fate of the core debris. Sich [4] has published an account of these investigations. The estimated distribution of the core debris within the damaged reactor is summarized in Table 3. These results indicate that 180 tons of the reactor fuel have been located. In light of the uncertainties in the mass determinations and the impossibility of exploring all parts of the reactor ruins, it appears that essentially all of the 190 tons of fuel have been located. Kisselev and Checherov [14] note, however, that the details of analyses and estimate of the reactor fuel distribution have not been published for independent examination. They also call attention to the complications posed by spent fuel in the reactor which could be confused for fuel dispersed from the reactor core. More recent investigations have been performed and updated figures of core debris distribution have been presented [39].

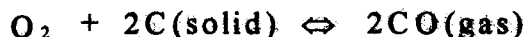
The most remarkable feature of the core debris is that some fraction of it melted and dissolved into the serpentine concrete lower biological shield of the reactor. This molten mass flowed into lower portions of the reactor where it froze. Apparently [4], little of the materials dropped from helicopters into the reactor vault actually landed on core debris. Consequently, these efforts at accident management little affected core debris behaviour or the releases of radioactive materials from the reactor.

Another remarkable finding of post-accident inspections of the reactor is that most of the graphite moderator has burned and disappeared. In 1986, it was thought that only about 20 percent of the graphite had burned, though some familiar with the damaged reactor claimed more had burned.

Based on these findings, Sich [4] has developed qualitative hypotheses concerning the behaviour of core debris following the initial dramatic events of the accident. He suggests that initially much of the fuel accumulated as a rubble bed intimately mixed with fragmented moderator graphite in the reactor vault. Combustion of the graphite in the rubble bed allowed fuel to segregate into an uncoolable mass. This mass heated sufficiently to penetrate into the lower biological shield. Eventually, a fairly fluid core debris-concrete mixture was able to flow out of the reactor vault and quench in lower regions of the reactor building. When this happened, radionuclide releases were

sharply reduced. Sharp rises in the radionuclide concentrations above the reactor on May 7-9 and May 16 may indicate core debris relocation events.

It appears that there is now sufficient evidence about core debris behaviour that predictions of the quantitative details of fuel behaviour including predictions of radionuclide release could be formulated. Following the initial, transient events, fuel would have been exposed to rather low oxygen potentials dictated by the equilibrium:



These conditions would favour the vaporization of radionuclides such as barium and strontium which are most volatile in the elemental state. Transport of vaporized species would be limited by the natural convection flow through a porous, gas-generating bed. As combustion of the graphite progressed, the porosity of the rubble bed would increase and fuel would be exposed to higher oxygen partial pressures. At higher oxygen partial pressures, radionuclides such as ruthenium, molybdenum, and technetium, which are most volatile in the oxidized state, would vaporize. Vaporization of the volatile oxides would become progressively less efficient as fuel segregated from the rubble of the moderator and attacked the biological shield at the base of the reactor vault. Partitioning of metallic fission products such as ruthenium and molybdenum from the fuel into the molten, steel liner of the biological shield would further affect vaporization of these elements. Exposure of the fuel to high partial pressures of oxygen from the air would be expected to cause oxidation to U_3O_8 . Strains produced in solid fuel by this oxidation will cause fracture and decrepitation. This fragmentation of the fuel by oxidation could be responsible for evidence of mechanical release of radionuclides late in the accident.

IV RADIONUCLIDE RELEASES

The fractional releases of individual radionuclides estimated to have occurred over the course of the Chernobyl accident in 1986 are shown in Table 4. The "initial estimate" in this table is that presented in 1986 by Soviet scientists [1]. The "current estimate" listed in the table is from reference 15 supplemented by comments by authors of this paper. The release of noble gases (krypton and xenon) was estimated to be 100 percent of the core inventory. The initial release estimates for the volatile radionuclides iodine, tellurium and cesium based on ground contamination in the Soviet Union varied between 10 and 20 percent. Releases of less volatile elements, ruthenium and molybdenum, were estimated to be between 2 and 3 percent. As discussed later in this paper, releases of ruthenium and molybdenum may even be higher than these early estimates. About 3 percent of the fuel and associated low volatility radionuclides were estimated to have been dispersed from the reactor building.

The apparently low releases of cesium and iodine were promptly explained. Initial release estimates shown in Table 4 were based on materials deposited only within the borders of the former Soviet Union. At the time the estimates shown in Table 4 were prepared, Soviet scientists did not have access to data on radionuclides deposited in other countries.

Warman [5] was among the first to publish revised, quantitative estimates of the Chernobyl reactor accident source term that included materials deposited outside the Soviet Union. Global dispersion codes (PATRIC, MESOS, GRID, ARAC) and compositions of samples obtained in various countries were used to estimate total iodine releases of 40-60 percent [18] of the initial core inventory. Tellurium and cesium releases were estimated to be between 30 and 50 percent of the initial core inventory. Confirmation of estimated releases of fuel and low volatility radionuclides could not be done by this technique since most of these materials deposited within the Soviet Union. A

more recent, more comprehensive analysis of the long-range transport of radionuclides outside the former Soviet Union has been published by Gudiksen et al [38]. They conclude release fractions for cesium and iodine were 40 and 60 %, respectively. They estimated the release fraction of tellurium to be only 10 %. Release fractions for other, condensable radionuclides are estimated to be small since most of these materials deposited within the Soviet Union relatively near the reactor site and were not included in their worldwide transport analysis.

Current, best estimate releases of radionuclides from the Chernobyl reactor have been prepared by Bedyayev et al. [15] and are listed in Table 4. The 33 % release fraction of cesium was first announced 1989 by A.A. Borovoi [40]. The cesium release fraction, 33 ± 10 percent, has also been suggested by Buzulukov and Dobrynin [13] and is consistent with the observed release of 47 percent of the cesium from residual fuel in the reactor and the observed cesium retention within the damaged reactor [15]. It is also consistent with analyses of the worldwide dispersion of cesium from the accident [5, 38]. Release fractions for iodine have been estimated to be as high as 80 percent, but the most likely range is between 50 and 60 percent. Tellurium release remains somewhat mysterious. Air sampling data in Nordic countries suggest that the release fraction for tellurium is at least as large as that for cesium and perhaps similar to that for iodine.

Estimated releases of barium and strontium could not be less than the releases estimated for low volatility elements such as cerium and zirconium, but reliable figures are not available on selective releases of strontium and barium that could augment the release fractions relative to those of non-volatile elements. Evidence from solubility measurements of particle samples taken at various distances from the Chernobyl site [34] indicates there was, indeed, some selective release of strontium.

From the earliest days of the Chernobyl accident, it has been known that many of the radioactive particles transported outside the former Soviet Union were nearly pure ruthenium [19]. The appearance of ruthenium particles was unexpected and the mechanism of release has been the subject of discussion [12, 20]. It is likely that ruthenium was released from the fuel as RuO_3 and RuO_4 vapors that condensed to form metallic particles. Kashparov et al. [35] suggest the reducing agents that caused the ruthenium oxides to be converted to ruthenium metal were particles from structural materials in the reactor.

One of the authors of the present report [21] has shown that, indeed, there were two periods of ruthenium release. The first was during the initial phase of the accident when mechanical, rather than chemical, processes were probably responsible for the release of substantial amounts of radionuclides as fine aerosol particles of fragmented fuel. The second phase of ruthenium release occurred several days later when it is possible fuel was exposed to high oxygen partial pressures and metallic inclusions in the fuel could react to form volatile ruthenium oxides. He argues that overall ruthenium releases and, perhaps, molybdenum releases are higher than any values shown in Table 4 because there was a sharp increase in the concentrations of ruthenium and molybdenum relative to concentrations of cerium and zirconium in air samples taken during later stages of radionuclide release [21]. The hypothesis of two phases of ruthenium release is consistent with the results of leaching tests by Kruglov et al. [34] that show there to be two physicochemical forms of ruthenium in particles deposited within the former Soviet Union. The relative abundance of the low solubility physicochemical form of ruthenium, which is presumably ruthenium in fuel fragments, decreases with distance from the Chernobyl site. The magnitudes of ruthenium and molybdenum releases cannot be accurately estimated except to note that they are higher than the initial estimates. Only 4 percent of the initial core inventory of ruthenium remains in damaged fuel in the reactor [17], but this could be indicative of both ruthenium release and partitioning of ruthenium into structural metals.

Releases of the low volatility elements such as cerium, zirconium, neptunium and plutonium are thought to have occurred by fuel fragmentation rather than by vaporization. Consequently, the release fractions for these low volatility elements are equal to the fraction of fragmented fuel dispersed from the reactor. As discussed further in the next section, this does not mean releases of the low volatility elements were confined to the initial, energetic, phase of the reactor accident. It appears mechanical release of fuel fragments persisted throughout much of the accident. Radiochemical analyses of land contamination of ^{144}Ce , which is expected to have vaporized little from the fuel, are interpreted now to indicate $3.5 \pm 0.5\%$ of the reactor fuel was dispersed from the plant as fragments.

V CHEMICAL AND PHYSICAL FORMS

Characterization of a severe accident source term includes description of the chemical forms of vapors and the physical forms of aerosol particles. Attention to these aspects of the Chernobyl source term have been based on samples of contaminated materials collected far from the reactor. Much attention has been devoted to the chemical form of iodine. The available data [21] show that much of the iodine was in gaseous form (I_2 or CH_3I). The fraction of iodine that was gaseous varied. Devell reported the gaseous fraction of the iodine to be 60 to 80 percent during the two weeks after accident initiation [16]. Winkelmann et al. [22] report that in the later stages of the accident 40 percent of the iodine was associated with aerosol particles, 35 percent was gaseous elemental iodine, and 25 percent was organic iodide. With time, the organic iodide fraction increased. Cambrey et al. [23] found about 75 percent of the iodine that reached the United Kingdom was in gaseous form. There clearly is no close chemical association between cesium and iodine. Data by Georgi [24] show there to be an exchange between gaseous iodine and iodine associated with particles. Furthermore, based on the chemical form of ^{132}I produced by decay of ^{132}Te , the time constant for iodine reaction to form organic iodides in the atmosphere is about 1 day [22].

Cesium in the released material from the Chernobyl accident did not remain entirely in a water soluble form. Salbu [25] found that only 35 percent of cesium in water was present as cations. The rest was bound to colloidal particles. Certainly, by the time the radioactive material reached Salbu's location (Norway), cesium was no longer present entirely as CsOH or CsI . This alteration of cesium to an insoluble form as well as the alteration of iodine from a particulate to a gaseous form may have generically applicable implications concerning the consequences of radionuclide dispersal into the environment.

One of the authors [21] has reviewed that state of understanding concerning aerosol particles produced by the Chernobyl accident. Again, much of the published information has come from samples collected in the West or particles collected from ground deposits. As a consequence, there is little known about size distributions, compositions, and the like for particles as they were emitted from the reactor. Particles that have been studied [19, 22-37] fall into three classes:

- o fuel fragments that contain fission products but have been depleted of volatile species such as iodine, cesium, and ruthenium;
- o mono-element particles; and
- o particles of condensed volatile radionuclides including cesium, tellurium and iodine.

Fuel fragments were certainly formed and lofted from the reactor building during the initial, energetic events of Chernobyl accident. Larger fragments ($>50\ \mu\text{m}$) were deposited near the Chernobyl site. Smaller fragments ($<20\ \mu\text{m}$) were carried well beyond the site and even beyond the bor-

ders of the former Soviet Union and have been detected in Poland [36], Greece [32], Hungary [37], as well as the Nordic countries. Interestingly, fragments have been found that include carbon which may have come from the reactor moderator [29].

Fragmentation of the fuel may have persisted throughout the accident. Hot particles consisting of fuel fragments have been detected in Greece which was not exposed to the energetic plume formed during the initial phase of the Chernobyl accident [32]. A possible mechanism for fuel fragmentation late in the accident is air oxidation of relatively cool ($< 1000^{\circ}\text{C}$) fuel. Air oxidation of uranium dioxide to form U_3O_8 is known to cause decrepitation. Fine fuel fragments, depleted in volatile radionuclides but still containing nearly the initial concentrations of low volatility radionuclides such as zirconium and cerium, could have been dispersed by the strong updraft known to have developed through the core. Certainly, Sich [4] notes that the entire Chernobyl site is heavily contaminated with dust particles he attributes to the oxidation and decrepitation of fuel

Kashparov et al. [35] have shown that aerosol particles with a bimodal size distribution are produced when Chernobyl fuel is heated to 673 to 1173 K in air. Size distribution data for these particles can be fit assuming two lognormal distributions with mean diameters of $5.6 \mu\text{m}$ ($s_g=1.21$) and $11.4 \mu\text{m}$ ($s_g=1.19$). The proportion of fine particles increases with the duration of heating. After about 16 hours of heating, more than 60 percent of the aerosol emissions were found to belong to the smaller size mode.

The test results obtained by Kashparov et al. certainly show that it is possible for fuel fragment release to have continued well after the initial core disruption event at the Chernobyl accident. That is, fuel fragments detected at later times outside the Soviet Union need not have been resuspended fragments produced by the initial energetic events of the accident. The relative amounts of fuel fragment release by the initial energetic events and subsequent reactive processes are not known.

Volatile fission products (cesium, iodine and tellurium) are found in very small particles ($0.5 - 1 \mu\text{m}$) that have compositions different than fuel. Jost et al. [28] find that iodine is associated with smaller particles than cesium, ruthenium, or tellurium. Georgi [24] observed a similar phenomenon.

VI CONCLUSIONS

A consensus is emerging concerning the general features of the Chernobyl accident and the releases of radioactive materials to the environment during this accident. Knowledge of the radioactive releases has advanced considerably since 1986 as a result of further analyses of environment samples taken worldwide as well as the results of further examinations of the destroyed reactor and analyses of core materials.

It can be concluded now that:

- o The release of radioactivity extended over more than a week with two pronounced, intense, emission periods. The first intense, emission period was associated with the accident initiation. The second emission period occurred about a week later when damaged reactor fuel may have been exposed to oxidizing conditions. The exact pattern of radioactive material releases between these two intense periods is not known. About 9 or 10 days after accident initiation, releases of radioactivity fell to very low levels. There is information that suggests some minor peaks in radioactivity releases occurred some weeks later.
- o There is a better understanding of the total release of radioactivity to the environment. Releases consisted of gases, vapors, aerosols, fragmented and probably reacted fuel. Current best estimates of the release fractions of various isotopes (see Table 4) are somewhat different than previous estimates and may be revised further in the future.
- o More than half the initial core inventory of iodine is thought to have been released. As a result of integration of worldwide deposition and analyses of core debris within the reactor, the release of cesium is thought to amount to about one-third the initial core inventory.
- o A peculiar feature of the radioactive material release during the Chernobyl accident is the release of a substantial amount (3.5 percent) of the fuel to the environment as fragments. Release of fuel as fragments occurred during both periods of intense release of radioactivity. Release of fuel fragments late in the accident may be the result of core debris oxidation. Low volatility elements such as cerium, zirconium and the actinides were retained in the fuel fragments and little selective release of these elements by vaporization can be detected. Fuel fragments deposited faster and closer to the reactor site than did cesium and iodine but were detected in Poland, the Nordic countries and Greece.
- o Another unexpected feature of radionuclide release during the Chernobyl accident was the appearance of particles composed almost totally of ruthenium isotopes. The probable mechanism for the formation of these particles is the vaporization of ruthenium oxides from the fuel and the subsequent condensation and reduction of the vapors to form metallic particles.
- o Air samples indicate that the releases of molybdenum and ruthenium were substantially higher in the late phase of the radionuclide emission period than were releases of fuel fragments which suggests selective release of molybdenum and ruthenium by oxidation to volatile forms.

- o The composition and the characteristics of radioactive materials released from the reactor changed during transport due to gravitational settling, wet and dry deposition, and chemical transformations. Chemical transformation of released materials such as the formation of gaseous iodine and water-insoluble forms of cesium could have generic implications for the estimation of the consequences of nuclear reactor accidents.

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Prof. A.A. Borovoi at the Russian Research Centre, Kurchatov Institute, as well as many colleagues within OECD countries, have examined the report and suggested modifications. The authors would like to express their sincere thanks to all of them.

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Table 1. Estimated Burnups of Fuel Assemblies in the Chernobyl Unit 4 Core [3]

Number of Fuel Assemblies	Burnup Range (MWe/t)
721	14766 to 13029
392	13029 to 11292
154	11292 to 9554
101	9554 to 7817
35	7817 to 6080
43	6080 to 4343
41	4343 to 2606
172	2606 to 0
Average	10912

Table 2. Estimates of Radionuclide Inventories*

Radionuclide	Inventory (Bq) by							Begichev et al. [3]
	Warman et al. [5]	Clough [7]	Autilia [6]	Kirchner & Noack [9]	Little [10]	Sich [4]		
⁸⁵ Kr	3.3x10 ¹⁶	--	--	--	2.5x10 ¹⁶	2.8x10 ¹⁶	3.3x10 ¹⁶	
¹³³ Xe	7.3x10 ¹⁸	--	--	--	6.2x10 ¹⁸	6.5x10 ¹⁸	6.3x10 ¹⁸	
¹³¹ I	3.1x10 ¹⁸	2.9x10 ¹⁸	2.9x10 ¹⁸	2.4x10 ¹⁸	3.0x10 ¹⁸	3.1x10 ¹⁸	3.2x10 ¹⁸	
¹³⁴ Cs	1.9x10 ¹⁷	1.1x10 ¹⁷	1.6x10 ¹⁷	1.4x10 ¹⁷	2.0x10 ¹⁷	1.7x10 ¹⁷	1.8x10 ¹⁷	
¹³⁶ Cs	--	--	--	9.0x10 ¹⁶	9.6x10 ¹⁶	6.3x10 ¹⁸ (1.1 x 10 ¹⁷)	--	
¹³⁷ Cs	2.9x10 ¹⁷	2.4x10 ¹⁷	2.2x10 ¹⁷	2.7x10 ¹⁷	2.3x10 ¹⁷	2.6x10 ¹⁷	2.8x10 ¹⁷	
¹³² Te	3.3x10 ¹⁸	4.1x10 ¹⁸	4.4x10 ¹⁸	4.4x10 ¹⁸	4.1x10 ¹⁸	4.5x10 ¹⁸	2.7x10 ¹⁸	
⁸⁹ Sr	2.3x10 ¹⁸	3.6x10 ¹⁸	4.0x10 ¹⁸	3.2x10 ¹⁸	3.0x10 ¹⁸	4.0x10 ¹⁸	2.3x10 ¹⁸	
⁹⁰ Sr	2.0x10 ¹⁷	2.0x10 ¹⁷	1.8x10 ¹⁷	2.0x10 ¹⁷	1.7x10 ¹⁷	2.3x10 ¹⁷	2.0x10 ¹⁷	
¹⁴⁰ Ba	5.3x10 ¹⁸	5.8x10 ¹⁸	5.6x10 ¹⁸	5.5x10 ¹⁸	5.4x10 ¹⁸	6.1x10 ¹⁸	4.8x10 ¹⁸	
⁹⁵ Zr	--	5.8x10 ¹⁸	--	5.3x10 ¹⁸	5.1x10 ¹⁸	5.8x10 ¹⁸	5.6x10 ¹⁸	
⁹⁹ Mo	7.3x10 ¹⁹ (7.3x10 ¹⁸)	5.5x10 ¹⁸	5.7x10 ¹⁸	--	5.2x10 ¹⁸	6.1x10 ¹⁸	4.8x10 ¹⁸	
¹⁰³ Ru	5.0x10 ¹⁸	4.3x10 ¹⁸	4.0x10 ¹⁸	4.6x10 ¹⁸	4.5x10 ¹⁸	3.8x10 ¹⁸	4.8x10 ¹⁸	
¹⁰⁶ Ru	2.0x10 ¹⁸	8.9x10 ¹⁷	7.9x10 ¹⁷	1.1x10 ¹⁸	1.2x10 ¹⁸	8.6x10 ¹⁷	2.1x10 ¹⁸	
¹⁴¹ Ce	5.6x10 ¹⁸	5.6x10 ¹⁸	5.4x10 ¹⁸	--	5.1x10 ¹⁸	5.6x10 ¹⁸	5.6x10 ¹⁸	
¹⁴⁴ Ce	3.2x10 ¹⁸	3.9x10 ¹⁸	3.4x10 ¹⁸	3.8x10 ¹⁸	3.4x10 ¹⁸	3.9x10 ¹⁸	3.3x10 ¹⁸	
²³⁸ Pu	1.0x10 ¹⁵	--	--	7.3x10 ¹⁴	1.6x10 ¹⁵	1.3x10 ¹⁵	1.0x10 ¹⁵	
²³⁹ Pu	8.5x10 ¹⁴	--	--	8.0x10 ¹⁴	9.6x10 ¹⁴	9.5x10 ¹⁴	8.5x10 ¹⁴	
²⁴⁰ Pu	1.2x10 ¹⁵	--	--	1.6x10 ¹⁵	1.6x10 ¹⁵	1.5x10 ¹⁵	1.2x10 ¹⁵	
²⁴¹ Pu	1.7x10 ¹⁷	--	--	1.9x10 ¹⁷	1.8x10 ¹⁷	1.8x10 ¹⁷	1.7x10 ¹⁷	
²³⁹ Np	3.6x10 ¹⁸ (3.6x10 ¹⁹)	4.7x10 ¹⁹	5.1x10 ¹⁶	6.1x10 ¹⁹	6.7x10 ¹⁹	5.8x10 ¹⁹	2.7x10 ¹⁹	
²⁴² Cm	2.5x10 ¹⁶	--	--	3.3x10 ¹⁶	3.3x10 ¹⁶	4.3x10 ¹⁶	2.6x10 ¹⁹	

Footnote *:
at the time of accident initiation. Figures within brackets are final figures after correction. Short lived I 132 and I 133 inventories were 1.4 and 2.1 times that of I 131 respectively.

Table 3. Core Debris Locations [4]

Location	Fuel Mass (metric tons)
Upper Level of the Reactor Building	37.7 ± 5
Subreactor Region	93.7 ± 26
Steam Distribution Corridor	27 ± 8
Pressure Suppression Levels	12.5 ± 3
Estimated Fuel Released Beyond Station	6.7 ± 1
Total	180 ± 28

Table 4. Estimates of Radionuclide Releases During the Chernobyl Accident

Radionuclide	Percent of Core Inventory Released	
	Initial Estimate [1] ^(a)	Current Estimate [15] ^(b)
⁸⁵ Kr	100	100
¹³³ Xe	100	100
¹³¹ I	20	50-60
¹³² Te	15	25-60 (c)
¹³⁴ Cs	10	33+10
¹³⁷ Cs	13	33+10
¹⁴⁰ Ba	5.6	4-6 (d)
⁹⁵ Zr	3.2	3.5
⁹⁹ Mo	2.3	(e)
¹⁰³ Ru	2.9	(e)
¹⁰⁶ Ru	2.9	(e)
¹⁴¹ Ce	2.3	3.5
¹⁴⁴ Ce	2.8	3.5
⁸⁹ Sr	4	4-6 (d)
⁹⁰ Sr	4	4-6 (d)
²³⁹ Np	3	3.5
²³⁸ Pu	3	3.5
²³⁹ Pu	3	3.5
²⁴⁰ Pu	3	3.5
²⁴¹ Pu	3	3.5
²⁴² Cm	3	3.5

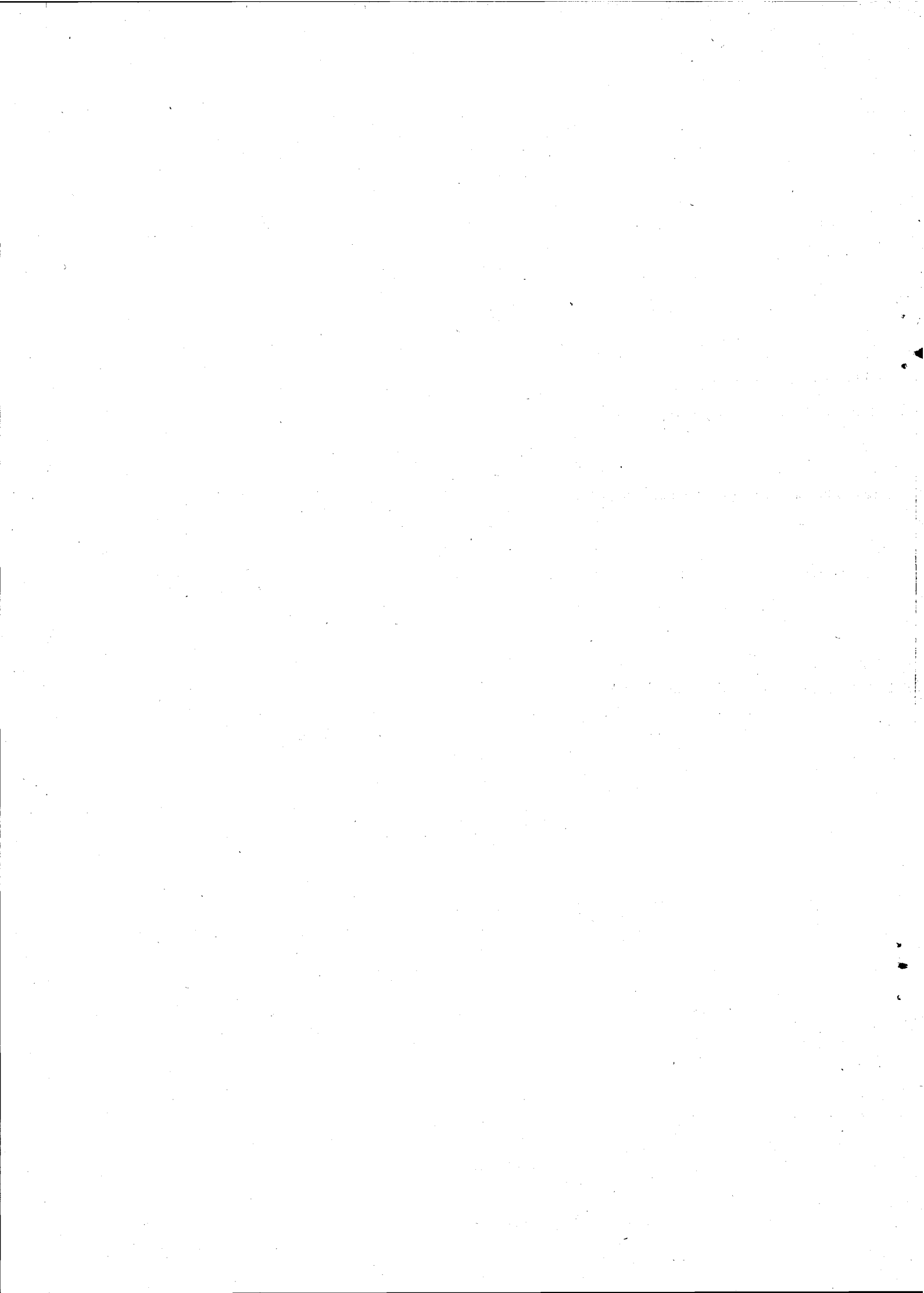
(a) Based on integration of deposited materials within the borders of the Soviet Union only.

(b) Comments by authors of this paper given in notes c-e.

(c) Air samples above the reactor and in Nordic countries indicate a release fraction 1 to 2 times that of cesium.

(d) Selective releases of strontium and barium in addition to releases caused by fuel fragmentation have been detected. Upper limits on these releases are uncertain.

(e) Probably substantially higher than the release of fuel material (~ 3.5 %) according to air samples [16] and depletion of radionuclides in core debris [17]. Air samples in Nordic countries indicate extensive Mo and Ru release relative to Ce, Zr, etc., release in the late phases of the accident



CURRENT EVALUATION OF THE CHERNOBYL REACTOR ACCIDENT RELEASE

A TECHNICAL OPINION PAPER PREPARED BY THE PRINCIPAL WORKING GROUP ON THE CONFINEMENT OF ACCIDENTAL RADIOACTIVE RELEASES (PWG-4) OF NEA's COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS (CSNI)*

Background

1996 will mark the tenth anniversary of the Chernobyl accident. The NEA Committee on Radiation Protection and Public Health (CRPPH) is preparing for that occasion an authoritative collective statement on the situation at Chernobyl, reviewing the current state of knowledge regarding the short-term and long-term health impact of the accident, discussing remaining problems (e.g., land contamination, population exposure), and presenting the lessons learned by the OECD Member countries in terms of radiation protection infrastructure and emergency preparedness. In order to make a full assessment of health effects, the CRPPH needed to consider dosimetric assessment. This implied, possibly, a re-evaluation of the magnitude of the Chernobyl accidental release. The CRPPH asked the Committee on the Safety of Nuclear Installations (CSNI) and the Principal Working Group on the Confinement of Accidental Radioactive Releases (PWG-4) to discuss this matter and come up, if necessary, with revised estimates for the release.

Preamble

The worst accident ever experienced at a nuclear power plant occurred on April 26, 1986 at Unit 4 of the Chernobyl Atomic Energy Station. This accident captured world-wide attention because the radioactive material released from the damaged plant was detected in most countries in the northern hemisphere. In August 1986, the scientists from the former Soviet Union provided the nuclear safety community at the IAEA Post-Accident Review Meeting in Vienna with a detailed account of the accident. This included preliminary assessments of the magnitudes, rates, and compositions of radionuclide releases (usually referred to as the "source term") during the ten days following initiation of the accident.

During the last decade, a great deal more data concerning the events, phenomena, processes and environmental consequences have been gathered after several investigations conducted at the Chernobyl site, and also at other places both inside and outside the former Soviet Union. As a result of the continued activities, the initiating event, the extent of the damage, the characterization of environmental release and its consequences have been, to an acceptable uncertainty, well understood. It is generally agreed that such an accident is highly unlikely in any western-type power reactor, and could not possibly lead in these plants to consequences as dramatic as those resulting from the Chernobyl accident.

The purpose of the following sections is to put the Chernobyl accident in perspective, very briefly, and to assemble and summarize the recent general findings and consensus concerning the Chernobyl source term.

* This paper makes use of the information presented on the previous pages.

Release Rates

The magnitude and timing of the release of radionuclides during the Chernobyl accident has been widely discussed over the last decade. There was an initial, intense phase of radioactive material release during the rapid reactor core disruption. The core disruption was due to the fast overpower transient caused by poor reactor operation and the very unfavourable reactor physics characteristics of the RBMK reactor concept. This initial release included fragments of fuel as well as other radioactive materials in the form of aerosol particles, gases and vapours. The simultaneous high energy release heated the plume of radioactive substances and lifted it high into the atmosphere. Much of the released material and especially the fragmented fuel particles deposited within the borders of the former USSR. Nevertheless, large quantities of radionuclides, especially the more volatile (e.g., cesium, iodine and tellurium isotopes), were carried large distances by high altitude air currents and were deposited across vast areas outside the former Soviet Union.

After the initial peak release during the core disruption phase, the releases of radionuclides continued but at a lower level, reaching a temporary minimum level around April 29, 1986. Large-scale efforts had been undertaken at the reactor site to mitigate and manage the accident; nevertheless, the release rate increased again. A second peak in the rate of radioactive material releases occurred. Finally, on or about May 5, some 9-10 days after the initial phase of the accident, the radioactive material release rates dropped by about a factor of thousand and have remained very low.

Release Composition and Magnitude

The initial quantitative estimates of the released radionuclides during the initial and subsequent phases of the accident presented at the Post-Accident Review Meeting in August 1986 were mainly based on the integration of ground deposits within the territories of the former Soviet Union. These have been upgraded to incorporate the data gathered on the radioactive material that fell out outside the former Soviet territories, and by closer examination of the destroyed reactor and analyses of materials from the core and from the reactor building. Clearly, the total release from the Chernobyl accident must have been greater than the material that fell within the former Soviet territories. Careful examination of the initial and currently known release estimates indicates that the total world-wide release of radiologically important nuclides, e.g. cesium and iodine, was 2 to 3 times more than what was initially estimated for the former Soviet Union alone.

More than half the initial core inventory of iodine is thought to have been released from the damaged reactor. As a result of integration of world-wide deposition and analyses of core debris within the reactor, one-third of the initial core inventories of cesium and tellurium are estimated to have been released.

Thus, for these materials, the total amount that was deposited over the whole world outside the former Soviet Union was of the same order as the amount that was deposited within the USSR. However, for other important materials, such as fuel fragments, relatively little was transported outside the former Soviet Union.

An exceptional feature of the radioactive material release during the Chernobyl accident was the release of a relatively substantial amount (3.5 percent) of the total fuel inventory to the environment. Release of fuel fragments late in the accident might be the result of core debris oxidation. Low volatility elements such as cerium, zirconium and the actinides were retained in the fuel fragments. Significant quantities of these elements outside of fuel particles were not detected. This implies that there was little release of these elements by vaporization. Certain radioisotopes within the fuel probably became liberated from the molten fuel or fuel fragments as a result of the high temperature and chemical environment in the days following the accident. This is probably the source of the particles composed almost totally of ruthenium isotopes. Air

samples indicated that the releases of molybdenum and ruthenium were substantially higher in the late phase of the radionuclide emission period than the releases of fuel fragments. This suggests that the release mechanism of molybdenum and ruthenium was by oxidation to volatile forms.

Although fuel fragments themselves were not carried out very far, monitoring at many locations showed that the fission products in gaseous or aerosol form were transported over large distances from the site. Distance tends to dilute the concentration of fission products in the atmosphere. However, climatic and local weather conditions often resulted in local concentration onto relatively small areas, even at large distances.

Conclusions

At the request of CRPPH, CSNI's PWG-4 re-evaluated the Chernobyl reactor accident release. The initial release estimates made by the Soviets in 1986 - based on limited data - did not cover territories outside the USSR. Subsequent studies conducted over the last decade have considered the additional results obtained from experts in Russia and other countries. The PWG-4 work has updated the release estimates making use of the more recent results. However, its re-evaluation does not modify the essence of earlier conclusions regarding the source term or health consequences; the latter will be discussed separately in documents prepared by the CRPPH.

