Paper 17

Electro-thermal phosphorus production, Radioactivity in the environment and workplace Justification, optimisation and alara considerations

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Summary:

The phosphate ore processed by Thermphos International in Vlissingen to produce elemental phosphorus contains approximately 1 Bq per g uranium 238 with daughter nuclides in near equilibrium (^{238}U+).

During the production process, radionuclides are emitted into the environment.

Slag is formed as a by-product. This is used as a construction material in road and hydraulic engineering, and causes a slight increase in external radiation. The process produces calcined precipitator dust, which has to be disposed of as radioactive waste. Operators concerned with the production of phosphorus receive a dose due to inhalation of radionuclides. This paper deals with the Justification of the process, the Optimisation measures and the application of the ALARA considerations to the individual and collective dose resulting from phosphorus production.

CONTENTS

1 General background and history

Hoechst Holland N.V. (referred to below as HHNV) began to produce elemental phosphorus from phosphate ore in April 1968. Since December 1971 three phosphorus furnaces have been in operation. Elemental phosphorus is used to produce a high-grade (thermal) phosphoric acid. As well as the phosphorus division, installations have been built at the Vlissingen site for the production of DMT, TAED and alkane sulphonate and for supplying energy. In July 1997 HHNV was divided into several independent companies. Thermphos International BV (referred to below as TIBV) is continuing the phosphorus production. For readability reasons, in this paper the name HHNV is replaced by TIBV as the legal successor carrying on the phosphorus production of HHNV.

In March 1983 the monitoring network, situated in the Sloe area to watch over the nearby nuclear power plant, detected radionuclides that appeared to come from TIBV. This was discussed for the first time with government representatives in May 1983. This meeting resulted in a request, which was submitted on 5 July 1985, for a licence under the Dutch Nuclear Power Act. The first such licence was granted on 30 December 1985. In 1984 the RIVM carried out emission/dose calculations. A recalculation took place in 1988.

In 1993 KEMA carried out another recalculation, based on the latest policy principles propagated by VROM (the Dutch Ministry of Public Housing, Planning and the Environment). After publication of the risk management policy, as applied by VROM to radiation, TIBV applied for an updated licence in 1994. The licence was granted on 28 December 1994.

The problem of radioactivity covers both the environment and the workplace.

2 The production process

2.1 Introduction

The phosphorus production process consists of various stages, which are described below in sequence.

2.2 Sintering plant

The phosphate ore is milled to a fine powder in this plant. The powder is brought onto a rotating granulator disk, together with a binder (clay suspension). Due to the rotation of the disk granules (pellets) are formed. The unsintered pellets are transported onto the front end of the slowly rotating sintering grid roaster. They pass through a drying zone (temperature up to 300 ^{o}C) and are then sintered to hard spheres under two large burners at temperatures around 800^{o}C. The pellets then pass through a cooling zone. The heat that they release is re-used in the drying zone.

The pellets are then conveyed to an intermediate storage facility, where they are stored in large silos before being fed into the electric furnaces.

There are three sintering roasters.

2.3 Slurry station

In the slurry station clay is suspended in water to produce the clay suspension used for granulation. Furthermore, precipitator dust, a return flow from the electric furnaces, is also added to the binding suspension.

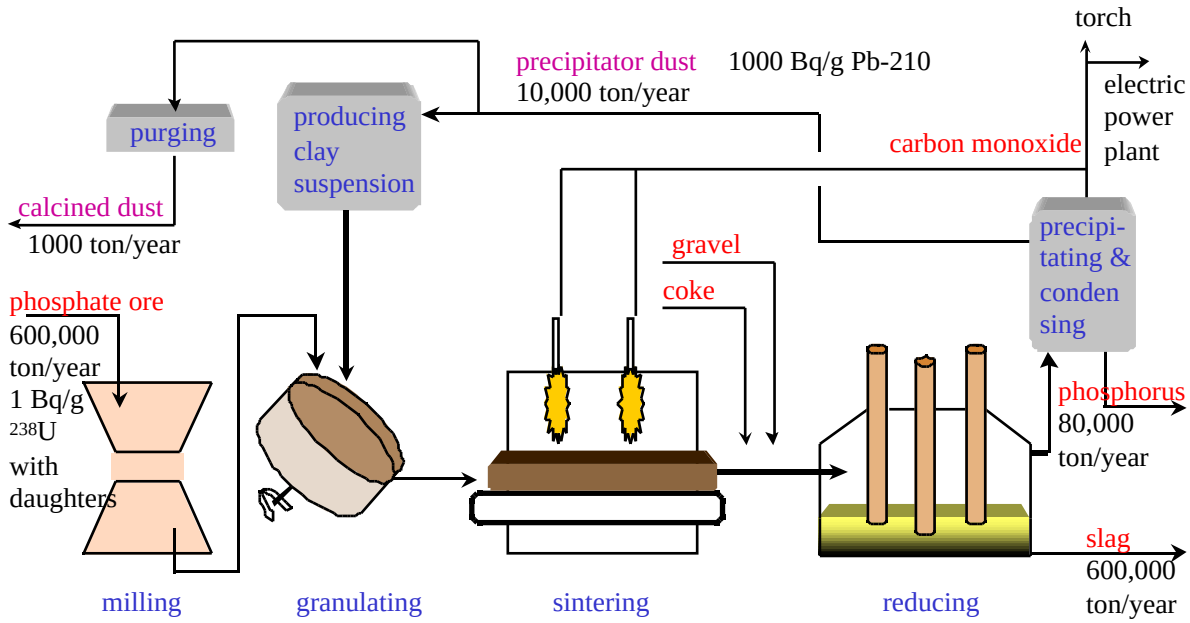
There are two slurry lines.

2.4 Purging unit

The slurry station also houses the purging unit. This is the discharge from the precipitator dust cycle that is formed in the production process (see 3.2). By discharging more or less of the precipitator dust, the concentration of volatile inorganic matter, metals and radionuclides in the precipitator dust cycle can be controlled. The purging unit consists of a high-pressure filter and a rotating calcining kiln where the pressed, lumped precipitator dust is calcined at temperatures of up to 750 ^{o}C.

2.5 Phosphorus plant

There are three electrothermal phosphorus furnaces in the phosphorus plant. The pellets are fed into the furnaces together with gravel and coke. In each furnace there are three electrodes, which draw electrical energy from a triangular arrangement of transformers and channel it into the furnace for heating the furnace contents. At temperatures higher than 1500°C a reaction takes place, in which phosphate ore is reduced to elemental phosphorus (P_4). The chemical reaction is shown in Figure 1.



$$2Ca_3(PO_4)_2 + 6SiO_2 + 10C \Rightarrow 6CaSiO_3 + 10CO + P_4$$

phosphate ore and gravel and coke => slag and carbon monoxide and phosphorus

PHOSPHORUS PRODUCTION OF THERMPHOS INTERNATIONAL

Figure 1

A calcium silicate slag is formed, which flows continuously from the furnace. The elemental phosphorus leaves the furnace as a gas, together with the carbon monoxide formed during the reaction. Entrained dust is separated from the gases in an electrostatic precipitator. This dust is collected in the slurry tanks, where it is mixed with water. The resulting precipitator slurry is pumped to the slurry station, where it is re-used in the granulator binder. The now dust-free gases are subsequently cooled, causing the phosphorus to condense to a stream of liquid phosphorus. The carbon monoxide (CO) is used as fuel in the sintering roaster. The excess gas is compressed and piped to a nearby electric power plant as a fuel.

3 Radioactivity problem

3.1 Introduction

The added mix of sedimentary and magmatic phosphate ore contains approximately 1 Bq per g of uranium with atomic number 238 (^{238}U). This nuclide has a half-life of $4.4*10^9$ years. All daughters of the ^{238}U decay chain are in a state of (approximate) equilibrium with the parent nuclide. This means that all daughter nuclides (see Figure 2) are also present with an activity of 1 Bq per g.

3.2 Enrichment mechanism

During the electro-thermal phosphorus production process, the radionuclides are unintentionally enriched.

At the high temperatures prevailing in the furnace, volatile inorganic substances, metals and radionuclides evaporate and condense on dust particles. The dust is trapped in the electrostatic precipitators and is recycled via the clay suspension into the pellets. When they reach the furnace for the second time, the volatile inorganic substances, metals and radionuclides evaporate again. In this way these substances are enriched in the so-called precipitator dust cycle. High concentrations of volatile inorganic matter and metals cause instability in the operation of the furnaces. To control the concentration the system is purged. The purge unit produces calcined dust with radionuclides and is regarded as radioactive waste under existing legislation (see 2.4).

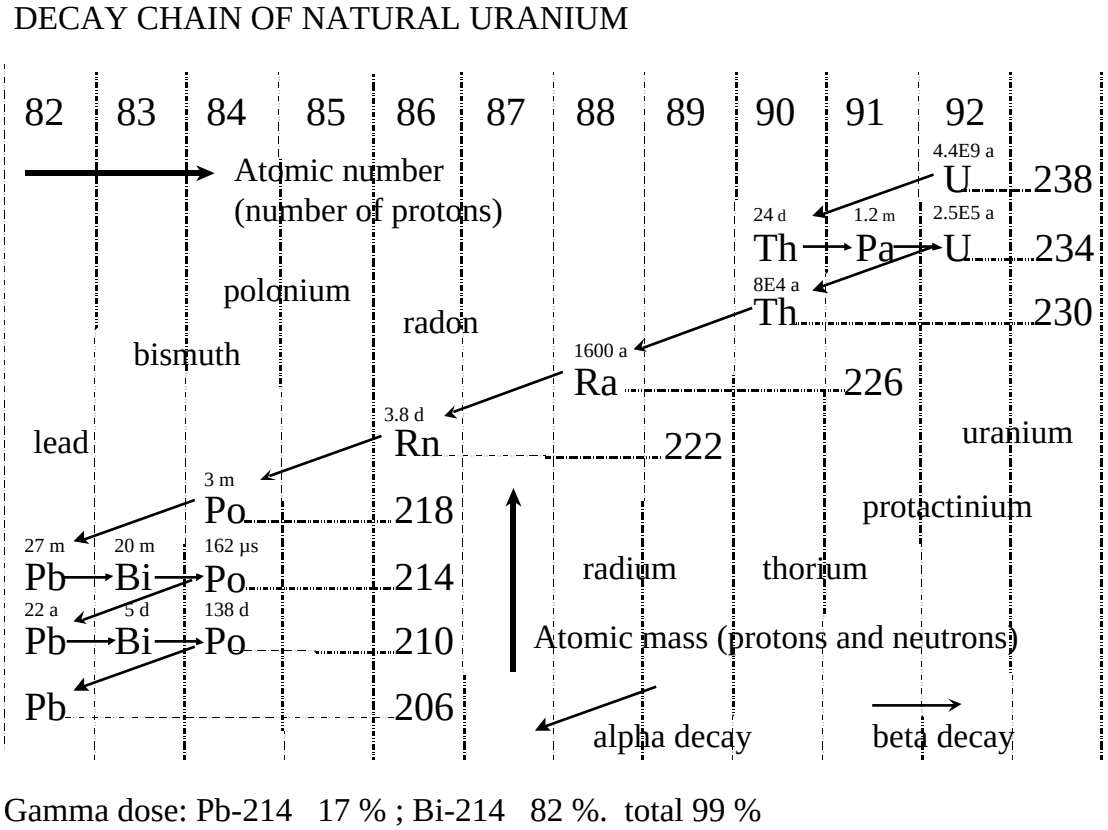


Figure 2

3.3 The uranium 238 chain (see Figure 2)

The radionuclides with an atomic number greater than that of radon remain in the furnace and therefore become part of the slag. Approximately one ton of slag is formed per ton of phosphate ore. Due to this fact the natural radionuclides in the ore are not enriched in the slag. Radon is an inert gas and therefore escapes along with carbon monoxide. The nuclides with an atomic number less than that of radon escape from the melt and therefore end up in the precipitator dust cycle. The short-lived nuclides with atomic numbers of 214 and 218 decay within a few hours. The problem nuclide is ^{210}Pb (lead), which has a half-life of 22 years and is therefore relatively long-lived. Moreover, most of the ^{210}Pb remains in the pellets in the sintering roaster. ^{210}Po, with a half-life of 138 days, is more volatile then ^{210}Pb and therefore leaves the pellets during the sintering process, passes both scrubbers (see 4.1) where the sinter gases are washed and is emitted into the environment. Consequently the activity of ^{210}Po in the precipitator dust is lower then the activity of ^{210}Pb.

The daughter nuclides formed from ^{210}Pb, {^{210}Bi (bismuth) and ^{210}Po (polonium)} have half-life times of 5 and 138 days. This means that, after about 4 half-life periods, these daughter nuclides exhibit nearly the same activity as the parent nuclide ^{210}Pb.

The activity of ^{210}Pb in the precipitator dust and in the calcined dust is approximately 1000 Bq per g. This nuclide has therefore been enriched by a factor of 1000.

3.4 Recognition of the problem

From the very start of TIBV in Vlissingen it was known that the phosphate ore contained enhanced concentrations of radionuclides. The problem of enrichment of radioactivity and the associated emission was not recognised until 1983, when the monitoring network of the nearby nuclear power station detected activity emitted by TIBV.

3.5 Measurement effort

TIBV's own environmental sampling team takes 200 samples of the air and water discharges each year.

TIBV has set up a C-laboratory to enable the emissions of radionuclides to be determined. The measurements needed to check for compliance with the environmental and workplace regulations, imposed by the Nuclear Power Act licence, are carried out here.

See Table 1 for the emissions during the period from 1987 to 1997.

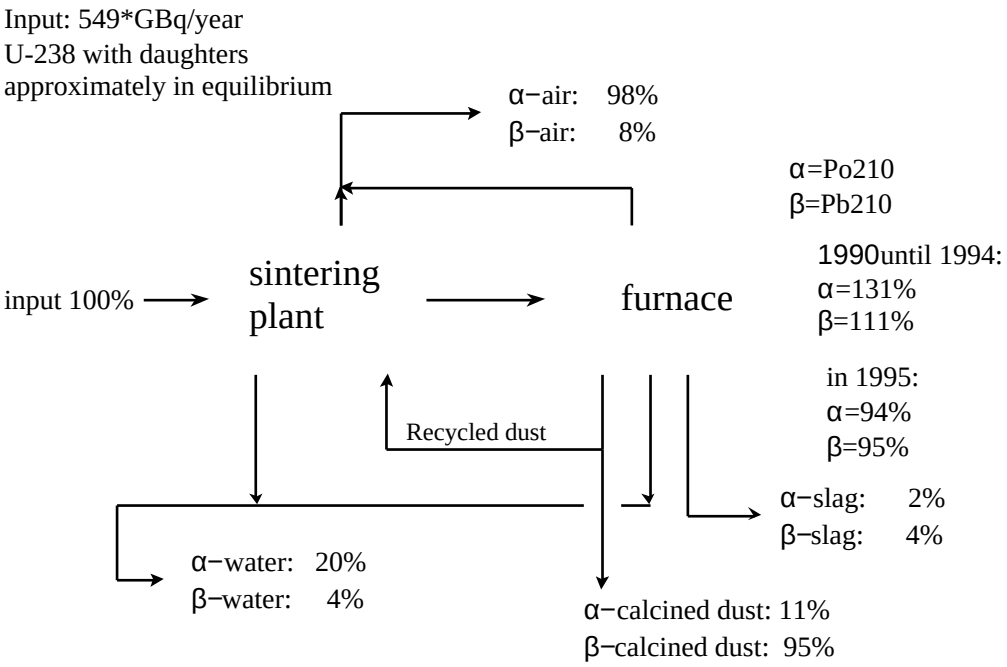
Table 1 Emissions to air and water

Year	_ to air ^{210}Po GBq/y	_ to air ^{210}Pb GBq/y	_ to water ^{210}Po GBq/y	_ to water ^{210}Pb GBq/y
1987	538	50	73	69
1988	843	98	95	40
1989	634	50	99	34
1990	381	34	107	24
1991	687	32	91	21
1992	490	66	166	24
1993	616	52	96	24
1994	506	33	82	29
1995	503	48	76	23
1996	390	95	58	36
1997	306	106	29	21
Average	536	60	88	31

4 Emission and balance

4.1 Emission and balance

The major emitter of radionuclides is the sintering plant. In 1985 new seawater scrubbers were installed in sintering plants 1 and 2, downstream of the existing fresh-water scrubbers, for the purpose of reducing emissions, including radionuclides, into the environment. Despite the very intensive scrubbing of the sintering waste gases, emissions of radionuclides into the environment occur. Figure 3 shows a summary of the emission balance.



Based on the period 1990 until 1994

Figure 3

In general it can be said that the volatile ^{210}Po is emitted almost in its entirety, while the greater part of ^{210}Pb ends up in the calcined precipitator dust. More is measured than is assumed as input to the process.

This was explained in 1994 in terms of:

o measurement accuracy;
o unknown variations in the phosphate ore;
o non-inclusion of the activity in the gravel and coke input;
o the formation of ^{210}Po from the ^{210}Pb in circulation, which can result in more ^{210}Po being emitted than was present in the input to the process.

The activity of all phosphate ores used during 1995 was measured. The measurements give higher concentrations of radionuclides in the ore then assumed in the years before. The result for 1995 is separately shown in figure 3 and is more realistic compared with the period 1990-1994. This emission balance counts for the period that a mixture of ore from Florida and Kovdor was used. During the use of ore from Jordan and Israel it turned out that the emission profile changed. TIBV is still studying the effects of different ores on the emission. From June 1998 ore from Florida was used again and it is expected that Florida ore will be used during the rest of 1998 and 1999.

5 Dose analysis at TIBV

5.1 Dose study by RIVM

The RIVM (Dutch National Institute for Public Health and Environment) carried out dose studies in the past (RIVM 841217, 1984 and RIVM 248103038, 1988). In order to obtain more insight into the actual dose, in 1991 the monitoring network that, in connection with the Borsele nuclear power station, had been installed in and around the Sloe area, was extended by a number of monitoring points, to enable the dose attributable to TIBV to be determined more accurately.

5.2 KEMA study

After consultation with VROM, TIBV commissioned KEMA to carry out a study in 1992 and 1993 to determine the dose caused by TIBV.

The dose to which the surrounding environment was exposed was calculated on the basis of the measured emissions over the period from 1987 to 1991 and the measured immissions in 1991.

A PC was connected to the wind vane on TIBV's office building for the purpose of recording the wind direction, speed and turbulence at hourly intervals. These data were used to determine the relationship between emission and immission.

The following exposure paths were examined:

o Inhalation of radioactive dust from discharges into the atmosphere.
o Ingestion of radioactive fall-out from discharges into the atmosphere:
- soil-plant-man
- soil-grass-cattle-milk-man
- soil-grass-cattle-meat-man
- water-shellfish/crustaceans-man
- water-fish-man

o Ingestion from radioactivity discharged directly into water:
- water-shellfish/crustacean-man
- water-fish-man

o External irradiation from cloud
o External irradiation from the soil, attributable to radioactive fall-out.

This study showed that the individual dose caused by TIBV in Nieuwdorp (a nearby village 4 km north-east of the production site) does not, on average, exceed 30 _Sv per year. The greatest dose is attributable to the inhalation path, which accounts for 96% of the total dose.

6 Policy principles of VROM 1994

6.1 Checking against policy principles

The situation at TIBV was checked against the VROM's POLICY PRINCIPLES RELATING TO RADIATION HYGIENE, version 15, December 1993. As well as a "24-hour exposure outside the site" dose, a MID (multifunctional individual dose) and an AID (actual individual dose) were introduced. If, for one practice or work activity, either the MID or the AID is below the threshold dose value of MTD of 40 _Sv per year, the ALARA considerations apply. This means that optimisation costs have to be weighed against the degree of dose reduction. If the MID and AID both exceed the MTD (40 _Sv per year), there is an "urgent remedial action" situation.

6.2 24-hour exposure outside the site

The applied policy requires calculation of the dose to which a person would be exposed during 24 hours at places (just) outside the plant (MID without the protection provided by housing: MID*). All possible exposure paths are examined (inhalation, ingestion and external gamma radiation).

6.3 Residential reduction factor and MID

The dose attributable to external radiation can be multiplied by 0.25 before it is added to the other doses in order to calculate the total dose. This is because residence is considered as the most common form of long-term sojourn. The protective action (effect) of a residence (building) is set at a factor of 4. This makes the MID a criterion for a "remedial action" situation. The MID must be smaller than the MTD of 40 _Sv per year, otherwise there is a "remedial action" situation. Then the AID can be examined to determine the urgency associated with the "remedial action" situation.

6.4 Sojourn factor and AID

The AID is determined by taking the MID without residential protection, and multiplying it by an ABC factor (actual exposure correction factor). The ABC factor is not applied to the ingestion path. If the AID is smaller than the MTD of 40 _Sv per year, the "remedial action" situation is not urgent.

6.5 Summary

Table 2 shows a summary of the calculations from the evaluation.

Table 2: Summary of evaluation against policy principles.
Note: The figures are based on measurements carried out during the period from 1987 to 1991.

Location Site-perimeter and village	Dose Conti-Nuous Inhala-Tion _Sv/y	Dose conti-nuous exter-nal radia-tion _Sv/y	MID* inhala-tion + exter-nal radia-tion _Sv/y	ABC factor	AID inhala-tion + inter-nal radia-tion _Sv/y	MID inges-tion _Sv/y	AID total _Sv/y
Van Cittershaven	112	29	141	0.01	1.4	1.2	2.6
SE corner of site	76	1.7	78	0.01	0.78	1.2	2
Aquila	52	7.7	60	0.01	0.6	1.2	1.8
ELF-Atochem	102	5.4	108	0.2	21.6	1.2	22.7
P&H	147	2.6	150	0.2	30	1.2	31.1
Pechiney	141	1	142	0.2	28	1.2	29.6
Nieuwdorp	29	0.04	29	1	29	1.2	30.2

*without the protection provided by housing.

6.6 Conclusions in 1994

The following conclusions can be drawn from the above:

- A "remedial action" situation does not exist in Nieuwdorp due to the MID* of 29 _Sv per year and the AID of 30.2 _Sv per year.
- There is not an "urgent remedial action" situation at the other locations (see AID total).
- The ALARA considerations must be applied at both locations, with optimisation costs being weighed against dose reduction.

6.7 Legislation and policy principles of VROM in 1996

In 1996 the Council of the European Union adopted a directive on the protection of the health of workers and members of the public against the dangers of ionising radiation. In this directive the cumulative maximum dose for members of the public is set at 1000 _Sv per year.

VROM decided in 1996 to affirm the MTD (maximum tolerable dose) for one practice or work activity at 100 _Sv per year and changed the Nuclear Power Act accordingly. As a result the Dutch legislation is less strict then the former policy principles (1994) and therefore more in agreement with the European Union directive. VROM based its support of the 100 _Sv per year threshold limit by arguing that it is possible that members of the public could be exposed to a maximum of 10 different practices or work activities. Another benefit of this approach is that one practice or work activity will not be dependent on another not filling up the licensed space to the maximum cumulative dose of 1000 _Sv per year.

The dose is still the prime unit for legislation instead of the previously used risk in the policy principles.

As a result of this changed legislation the conclusions under 6.6 are still valid in 1998.

6.8 Situation in 1998

TIBV is continuously studying the impact of phosphorus production on the environment. Together with scientist from KEMA there was a re-evaluation that took the new model of the human respiratory tract, described

in ICRP publication 66, into account. The AID in Nieuwdorp appeared to be 21 _Sv per year instead of the former 30.2 _Sv per year. There are still measurements going on, like the real particle size distribution occurring in the environment and the rate at which the radionuclides are leached out from the emitted dust, to improve the dose estimations. At this moment (September 1998) it is not expected that the conclusions under 6.6 will change.

6.9 Legislation and policy principles of VROM in 1998

The Dutch Government is implementing the new European directive. The European directive has an annex with exemption levels. A practice may be exempted from the requirement of reporting and/or licensing if the activity concentration or the quantity does not exceed the values in the annex. However, the exemption levels in the annex are intended to be used only for radionuclides that are processed because of their radioactive properties (the functional activity). Exemption levels for radionuclides in enhanced concentrations that are not processed because of their radioactive properties (the non-functional activity) are excluded from the prime scope of the directive. All the activity in the phosphorus production process must be regarded as non-functional. Exemption levels for these non-functional radionuclides are being developed now. It is not clear yet (September 1998) whether the material “slags used in road and dike building” will become subject to legislation. Similarly the exemption or clearance levels for calcined dust are not yet known. The storage time as a radioactive waste is determined directly by these exemption levels or (conditional) clearance levels. The European Union has attached a reporting obligation to the exemption levels. The consequences of this reporting obligation for the use of slag as a construction material are not clear because the exemption levels for non-functional radionuclides were not known when this article was written (September 1998).

The EU directive gives member states the possibility to apply parts of the directive intended for the practices (functional) to work activities (non-functional).

7 Justification of phosphorus production

7.1 Advantages of phosphorus production

There is an international market for phosphorus and derived products. Customers and consumers purchase the products because they benefit from the application or consumption of these products. TIBV contributes with the phosphorus production to the Dutch and European economy. TIBV employs 450 employees directly and approximately 500 people indirectly. A measure of the benefit to society could be taken to be the wages paid to these employees. In this way the benefit of the phosphorus production is determined as approximately NLG 75 million per year.

7.2 Disadvantages

Because phosphorus is produced from phosphate ore, which contains enhanced concentrations of natural radionuclides, members of the public receive an individual dose and a collective dose due to emissions. The individual dose caused by these emissions is within the limits set by legal standards. The level of collective dose caused by these emissions is such that the examined feasible optimisations cannot be justified by a Cost Benefit Analysis (CBA).

Another disadvantage is the dose and collective dose caused by the phosphorus slag used in road and hydraulic engineering applications. However, the activity concentration in the phosphorus slag is, under current law, below the level at which a licence for use is needed. It is expected that, after the implementation of the new European Directive, there will still be no need for licensing. The individual and collective dose caused by the building material “phosphorus slag” lies within the range of the variations caused by the natural background and the construction materials used in the house building sector. The application in road and dike constructions is an optimisation, in view of the collective dose that is avoided by not using the slag in the house-building sector. If the additional risk is ranked in generally “accepted” risks, it has a low level of significance.

7.3 Consideration of the advantages and disadvantages

The disadvantages such as the caused dose are within legal standards. Furthermore, the costs attributable to the caused collective dose as calculated by a CBA only amount to a fraction to the economic advantage. TIBV therefore is of the opinion that the advantages of the phosphorus production outweigh the disadvantages and therefore is of the opinion that the phosphorus production is justified. Furthermore, it is demonstrated that, using a CBA-analysis value to judge the feasible optimisation measures, from this point of view there are no terms for further optimisations. However, TIBV continuously investigates the possibilities to reduce the emissions occurring in the phosphorus production and the consequently caused dose (See 10.3).

8 Optimisation and alara considerations and phosphorus production

8.1 General

Because it has been found that the MID caused by TIBV is below the MTD threshold of 40 _Sv per year, a "remedial action" situation does not exist in Nieuwdorp. Moreover, because it has been determined that, at the measuring points around the site, the MID exceeds the MTD (40 _Sv per year), while the AID is below the MTD, a "non-urgent remedial action" situation exists here. An ALARA analysis has to be carried out. This means that the effects of measures, such as the precipitator dust processing plant, on the reduction of the dose in the surrounding area (in this case by roughly half) must be weighed against the investment costs of NLG 28 million and the operating costs of NLG 6 million/year over 25 years. The Dutch policy statements give no clear indication of what costs are defensible in relation to the reduction of individual or collective dose. In the absence of nationally or internationally accepted values, TIBV referred to a CBA value for the purpose of weighing up the reasonableness of an investment. This is a value that indicates what investment can be considered reasonable for the purpose of achieving a reduction in collective dose. The figures published by the National Radiation Protection Board in 1993 specify £20,000 per man Sv for the general public (Doc. NRPB,4 No.2 75-80 1993). TIBV therefore applied the NRPB Cost Benefit Analysis value (CBA) of £20,000 per saved man-sievert in order to weigh up the reasonableness of an investment.

9 Optimisation of phosphorus production

9.1 Trials

Trials have been carried out (including the installation of demister mats) with the aim of improving the existing scrubber systems, but the results were disappointing (in terms of reduction in emissions of radionuclides, etc.).

Moreover, extensive trials have been carried out to determine whether supplementary scrubber systems (venturi scrubbers) can bring about reductions in the emission of radionuclides. These scrubber systems are very expensive and are undesirable from an energy point of view (additional electrical power). Moreover, the filtration residue, namely dust with radionuclides, can, in view of its large volume and low activity, only be fed back into the precipitator dust cycle. As a consequence the effectiveness of this scrubbing is considerably reduced. *

After long and comprehensive research, it was found that only 2 dose-reducing measures are feasible: the construction of a precipitator dust processing plant and a high stack.

* Recent studies (1997 and 1998) even showed that more efficient scrubbing could lead to a higher dose. More efficient scrubbing first causes a removal of the bigger particles, which carry the radionuclides. In the end the activity which is not immobilised in the calcined dust, like ^{210}Po, leaves the stack on smaller particles, causing a higher dose due to increased lung deposition.

9.2 Precipitator dust processing plant

In order to ensure the stable operation of the furnace process, some of the precipitator dust must be removed from the precipitator dust cycle.

This precipitator dust is processed into calcined dust and is stored on site as radioactive waste. At the present time approximately 750 tons of precipitator dust are taken out of the process each year. This is the necessary amount for the process to proceed stably. Because storage of calcined dust is very expensive, only the absolutely necessary amount of precipitator dust is removed.

In the period 1990-1994 a pilot plant trial was carried out to determine whether the stored calcined dust and the precipitator dust could be chemically processed.

On the basis of this pilot plant, TIBV worked out plans for an industrial-scale precipitator dust and calcined dust processing plant (referred to below as a precipitator dust processing plant). In this plant the heavy metals, the radioactive fraction and the other substances are chemically separated from each other. A highly active lead sulphate fraction is formed, which has to be transported to COVRA (Centrale Organisatie voor Radioactief Afval: The Dutch central organisation that stores radioactive waste).

The advantages of the plant include the following:

oThe volume of radioactive waste would be reduced considerably (see reservation under “effectiveness” below).
o Due to the above reduction, it would be possible to remove more precipitator dust from the process. It would be possible to reduce the emission of radionuclides by a factor of about 2 if, instead of 1000 ton per year, 3000 ton per year were removed and processed in the precipitator dust processing plant. The level of dose in the surrounding area as a consequence of the emissions would also be reduced by a factor of 2.

The most important arguments against this plant are:

o Costs. The investment cost would be NLG 28,000,000 and the annual operating costs would be NLG 6,000,000.
o Safety. The concentration of radioactivity exposes the operators concerned to additional doses.
o Effectiveness. It cannot be excluded that the activity of the separated zinc fraction (150 ton per year with an expected activity of 30 Bq per g ^{210}Pb) will be above a future limiting value (10 Bq per g annex EU directive) and therefore be regarded as a radioactive waste. The most important objective of the project - concentration of activity - will then be only partly realised.
o Another major problem is the coupling with the STPP (phosphate in detergents) production. If this production should be terminated in Vlissingen at some future time, furnace acid (produced out of elemental phosphorus) would have to be used in place of wet-process phosphoric acid. This would involve a downward adjustment of the price of furnace acid to the price of phosphate ore, resulting in a cost item of NLG 14,000,000 per year.

9.3 Stack

The effect of building a new and higher stack was calculated in the KEMA study. The individual dose in Nieuwdorp would be roughly halved by building a 140 meter high stack.

9.4 Phosphate ore

The origin of the used ore determines the levels of impurities and radionuclides. These impurities are crucial for the emissions that occur during production. Changes in the combination of chemical impurities can either mobilise or immobilise other impurities like the radionuclides. The chemical mechanisms behind these phenomena are not really understood.

10 ALARA considerations and phosphorus production

10.1 Precipitator dust processing plant

The collective dose caused by TIBV is 2 man Sv per year. Based on an investment for 25 years, the collective dose reduction is:

2 [man Sv per year]*25 [years] /2 [reduction]= 25 man Sv. Applying the above mentioned CBA values, an investment of 25 [man Sv]*20.000 [£ per man Sv.]*2.79 [NLG per £] = NLG 1.4 million, can be regarded as defensible.

The investment required to build the precipitator dust processing plant is NLG 28 million. The operating costs would be 25 years * NLG 6 million per year = NLG 150 million, so that the total costs over 25 years would be NLG 178 million. This is many times higher than the sum the NRPB regards as reasonable for reducing the collective dose. The necessity for building a precipitator dust processing plant in order to reduce the dose in the surrounding area is therefore not given. In early 1993, after consultation with VROM, it was decided that the plant would not be built.

10.2 Stack

The collective dose reduction that can be achieved by building a 140 meter high stack is 2 man Sv per

year. Due to the applied calculation specifications in the policy principles, with a cut-off dose for optimising purposes of 4 _Sv per year, these 2 man Sv per year are an overestimate of the real reduction. On the basis of an investment for 25 years and the overestimated dose reduction of 2 man Sv per year, the reasonableness consideration indicates that a sum of NLG 2.8 million is defensible.

(2 [man Sv per year]*25 [years]*20.000 [£ per man Sv.]*2.79 [NLG per £] = NLG 2.8 million). The cost of a 140 meter high stack would be NLG 5 million. This sum, too, is higher than the sum that is regarded as defensible for a reduction in collective dose. Moreover the erection of a higher stack would not reduce emissions in absolute terms, so that the benefits would be restricted to the immediate vicinity. TIBV therefore decided, in consultation with VROM, not to build the stack.

10.3 Alternative ore

TIBV used predominantly ore from Florida until 1996 after which this ore was no longer available. For this reason Jordan ore was used in 1996 and 1997. The concentrations of the impurities in this ore differ from those in Florida ore. As a result of the differences in the ore, cadmium emissions to the environment increased. This was remarkable given the fact that the concentration of cadmium in the Jordan ore is lower than that in the Florida ore. In combination with other impurities, notably chloride or calcium carbonate, cadmium becomes volatile during sintering. In a similar way the ^{210}Pb emissions increased even though the concentrations were lower. The concentration of radionuclides in Jordan ore is about 30% of the concentration in Florida ore. During the period 1997 until July 1998 TIBV tested other ores and their influence on the emissions.

The choice of ore (origin) type is part of the ALARA considerations too. However, using unknown ores, their influence on emissions is not clear in advance, because of mobilising or immobilising characteristics. The emission profile belonging to new ore can only be determined after its use in the process during a period of up to several months.

The composition of the mixture of used ores is therefore determined by technical, economical and environmental conditions. Furthermore the long or short-term availability of ores on the world market influence this mixture.

TIBV stopped the use of Jordan ore because of the increased cadmium emissions. In the near future TIBV will evaluate this decision. ALARA considerations will be made with respect to the caused damage or risk of chemical and radiological compounds. The environment will be looked at as a whole and not as separated compartments.

11 Calcined dust

11.1 General

To enable the production process to proceed smoothly, approximately 750 tons of calcined dust has to be removed each year from the precipitator dust cycle. For a number of years this calcined dust has been kept in intermediate storage on site.

The calcined dust is classified as radioactive waste under current law (800 Bq per g ^{210}Pb+ $t^1/_2$=22 years) and therefore cannot be disposed of in a chemicals landfill.

11.2 Immobilisation

KEMA has carried out leaching tests, based on the Dutch government's limiting values memorandum "storten gevaarlijk afval" (dumping hazardous waste) of May 1993. The chemical composition of the waste is such that, after it has been immobilised in cement, it can be disposed of in a C3 landfill without any difficulties. The ^{210}Pb activity will have fallen below 10 Bq per g (exemption level for ^{210}Pb for functional radionuclides EU directive) after 150 years. After 150 years, or sooner or later if the (conditional) clearance levels for non-functional radionuclides are set at different levels in future, the calcined dust can be immobilised and disposed of in a C3 landfill.

11.3 Storage at the COVRA

TIBV and the COVRA have signed a contract to store the calcined precipitator dust.

A choice has been made for bulk storage rather than the standard packagings. The material will be stored in 20-foot containers with a plastic lining. These containers will be stacked in a purpose-built storage building.

Materials tests were carried out at KEMA. It has been decided that the calcined dust will not yet be immobilised, because immobilisation with cement will increase the volume. This is not desirable because the material will then take up more space in the storage building. Immobilisation can be carried out after the decay period of the radionuclides.

11.4 ALARA considerations and calcined dust

ALARA considerations do not apply to calcined dust. The general public receives no collective dose from this stored dust.

12 Workplace problems

12.1 Introduction

Operators who are involved in the production of phosphorus are exposed to radionuclides (^{210}Pb and ^{210}Po) predominantly from the precipitator dust cycle. These nuclides are alpha- and beta-emitters. To cause a dose, there must be an intake of these radionuclides into the human body. In workplaces an intake occurs predominantly by inhaling the dust that comes from the precipitator dust cycle.

12.2 Dust characteristics

The inhaled dust was examined by leaching tests in simulated lung moisture. The results of these tests showed that the relevant radionuclides (^{210}Pb and ^{210}Po) are leached out of the inhaled dust very poorly and as a consequence the lung retention class was set to class Y. This means that the radionuclides are retained in the lungs for periods of up to several years. Consequently the caused dose is received predominantly by the lungs.

12.3 Measurements

To obtain the dose received by the operators concerned, Personal Air Sample measurements were carried out.

The measurement device consists of an air pump and a filter in a filter holder, connected to each other by a tube. The measurement device is carried by the operator in such a way that a continuous sample of the air surrounding the operator is passed through the filter in the filter holder. The dust containing the radionuclides is separated from the air and collected on the filter. The operator carries the pump and filter with filter holder for the working period of 8 hours. The filter is measured for alpha- and beta-counts on a proportional counter tube. The intake of radionuclides can be calculated from the obtained count rate, the flow that passed through the filter and a worker's assumed breathing rate. Given the intake, together with a Dose Conversion Coefficient (Sv per Bq), it is possible to make a dose assessment.

12.4 Measurement effort and dose assessment

In the period 1984 until 1993 approximately 30 PAS-measurements per year were taken on the operators in working conditions in which inhalation of dust is most likely to occur. To learn more about specific working circumstances, if the dose assessment for a certain job exceeded a threshold value, more specific measurements were taken. By taking more measurements at the same time at different places in a working area and comparing them with earlier measurements, taking the working and process conditions into account, it was possible to determine the pathways by which the radionuclides from the process reach the operators involved. As a result, a number of measures were taken to prevent the inhalation of radionuclides in order to obtain dose reductions.

In general it can be said that operators concerned with the production of phosphorus at the TIBV site are exposed to an average dose of 1 mSv per year. This dose is not given in the same dose rate every day. There are days when the dose, extrapolated from one workday (the measuring period) to a year dose, yields an annual dose of 5 mSv per year. On the other hand, there are also days that yield an extrapolated dose of 0 mSv per year depending on the (determined) process and working conditions (pathways).

It is also found that it is possible to obtain dose rates up to 1 mSv per hour if parts of the installation, covered with high concentrations of radionuclides, are polished, jagged or subjected to other operations that remove radionuclides from the contaminated surface with a risk bringing them into the air.

Contamination up to several hundred Bq per cm^2 has been determined on surfaces on the inside of the process equipment. Operators are consequently, under normal circumstances, not exposed to these high levels of contamination. However, during plant stops or repair work, when the installation is opened, preventive measures should be taken to avoid a high inhalation dose.

12.5 Legislation

If workers can be exposed to a dose of more then 5 mSv per year, they should be regarded as radiological workers.

It is expected that this value will be decreased to 1 mSv per year by the year 2000. TIBV is in discussion with the competent authorities concerning whether it is necessary and appropriate to give the operators involved the status of radiological worker.

12.6 Improving measurements and system

Due to the expected stricter legislation, in combination with the implementation of the new "Human Respiratory Tract Model for Radiological Protection" by the ICRP, TIBV is improving the measurement methods for determining the dose operators are exposed to. In addition TIBV will make dose assessments for all the different functions involved in the production of phosphorus.

12.7 Optimisation and ALARA considerations and workplace

About 200 operators and technical staff work in the factories where the production of phosphorous takes place. Consequently the collective dose that this population receives in 25 years is 5 man Sv.

Assuming that an investment in improving working conditions would avert that collective dose, reasonable investment costs are about NLG 750,000, based on the CBA value of £50,000 per saved man Sv used for occupational situations.

The measures:

- oNew floors in the phosphorus plant that can be cleaned easily.
(The involved costs were NLG 600,000)
- oCentral vacuum cleaning system without the need to empty the filter system.
- oProcess automation to avoid work in which a dose is received.
- oBreathing air protection measures in situations where the creation of concentrations of radionuclides in the surrounding air is unavoidable.
- oContinuous cleaning operations carried out by several workers.

The dose reduction due to these improvements is not the complete 5 man Sv mentioned above but only a fraction of it. Therefore it is evident that the improvements were carried out are based on "good housekeeping" and "best technique available" considerations rather then on strict ALARA considerations.

13 Slag as a construction material

13.1 General

Slag ($CaSiO_3$) is a stone-like by-product and is used as a construction material in the hydraulic and road engineering sectors. The same material is found in nature and is extracted from the earth's crust for use.

Some of the benefits of using slag are:

- o Slag is a good substitute for gravel and prevents the formation of gravel pits caused by gravel mining.
- o The angular structure of the slag ensures that, using it in a road bed, the road is more resistant to subsidence. Together with steel-slag a hydraulic reaction occurs which makes the compound even more resistant to subsidence.

A disadvantage of using slag is that about 1 Bq per g ^{238}U is present in a state of equilibrium with its (gamma-emitting) daughters except ^{210}Pb and ^{210}Po. (Compared to the phosphate ore there is no enrichment of radionuclides in the slag. The phosphorus in the ore is replaced by silicon in the slag.)

The possible consequences of this are:

Distribution of radionuclides

Undesirable distribution of radionuclides in the environment which may cause an unacceptable dose to members of the public.

External irradiation

Due to the use of the slag in road and hydraulic engineering applications, members of the public could possibly be exposed to higher levels of background radiation especially by use in the house building sector.

13.2 Justification

Slag is an inevitable by-product of the phosphorus production.

Due to the large resulting volumes of the slag, an economically and environmentally viable use of slag is a precondition for the continuity of the phosphorus production.

Slag, as a construction material, should not be treated differently to other construction materials, which are allowed to cause doses in the range of several thousand _Sv per year. See 13.5.

In general, I doubt if the system of justification, optimisation and ALARA considerations should be applied to construction materials used in house, road and hydraulic engineering, as a practical way of dealing with the natural activity. In fact this is the reason why an alternative system is currently under consideration for the house building sector in the Netherlands. See 13.5.

Nevertheless, I will give you an estimation of the radiological facts: the caused dose and the related hypothetical risk of the slag and compare it with the dose caused by other building materials.

13.3 Optimisation of slag

If phosphorus slag is used in considerable quantities as construction material in a house it will cause a higher dose than comparable materials like gravel. To give an idea of the external radiation: staying for a year on a surface formed by a thick layer of phosphorus slag will cause a dose of approximately 5000 _Sv.

From the very start of its phosphorus production operations, TIBV therefore, in consultation with government bodies, has only released the slag for use in road and hydraulic engineering applications. Even 30 years ago, a system of “conditional clearance” was practised. Governmental bodies responsible for road and hydraulic engineering applications supported the use of the slag.

The top layer of a road surface, which never contains phosphorus slag, shields the external radiation emitted by the slag. Because the thickness of this top layer can vary, it is not possible to specify the exact dose rate associated with the use of slag in road building. In general terms it can be stated that the dose is of the order of magnitude of 40 _Sv per year for someone who spends 1 hour per day on a cycle path, and 40 _Sv per year for someone who spends 8 hours per day on the road. The difference in dose rate is attributable to the thickness of the top layer.

VROM commissioned ECN to carry out a study of the leaching behaviour of the radionuclides. The amounts that leach out appear to be so small that a hypothetical calculation - all the dikes of the IJsselmeer (the biggest lake in the Netherlands) were provided with slag as a filler material - yielded a maximum dose of 0.012 _Sv per year.

Calculations on slag:
The exact dose depends on a number of calculation factors that are not included in the policy principles (See 6.1):

- What sojourn times on roads and cycle paths should be assumed?
- What proportion of roads and cycle paths contain slag as construction material? Should the current situation or, for example, the situation in 25 years, be taken as the basis for the calculations?
- What background should be subtracted from the dose rate on the road? The soil at the side of the road or the average road without slag?

Other criteria may also be of importance in this context.

13.4 ALARA considerations and slag

Some figures for phosphorous slag:

- The production is 600,000 ton per year, sufficient to build a road with a length of 120 km.
- In the 30 years of our company’s existence, approximately 3500 km of roads could have been constructed with slag. This is approximately 3.5% of the Dutch road network.

oThe maximum dose caused by slag is no more then 40 _Sv per year. In practice it will be in the order of 1 _Sv per year because people stay on the road for an average of 2 hours daily rather than 8 hours and less than 4% of the roads are built with phosphorus slag.

oBased on a 2-hour daily sojourn on roads it can be calculated that the collective dose caused by the slag to 16 million people is 6 man Sv per year. With the NRPB CBA value an amount of NLG 350,000 per year could be spent to eliminate a dose of this magnitude. (If the slag was used for the house building sector for 30 years it would cause an estimated collective dose of 3500 man Sv.)

Not included in this consideration is the benefit of not using natural gravel resources and the fact that roads built with slag have more resistance to subsidence, which are also environmental and economic advantages for society. It may even be that these advantages are already sufficient to justify the use of slag apart from the advantages resulting from the phosphorus production. Construction and maintenance costs carried out on 3500 km of road are involved. The economic value of 1 ton of phosphorus slag as a construction material has been determined as NLG 22 (RIVM 771402022). TIBV and CE are studying in detail the benefits for society due to the use of slag.

The above calculations where made with rounded figures for a first estimation. As mentioned above (13.3) additional policy principles or legislation could necessitate adjustment to the figures.

13.5 Ranking the dose/risk

Another approach to judging the dose and risk attributable to the slag is to rank it with the “accepted” risks caused by other applications to put it in perspective.

Comparison with natural background.

The background radiation in the Netherlands is approximately 2000 _Sv per year of which 1500 _Sv per year are caused by house construction materials.

The collective dose caused by the slag is a fraction of the dose caused by the house building materials, which is approximately $16*10^6$ (inhabitants Netherlands)*1500 _Sv per year (dose per inhabitant)= 24,000 man-sievert per year.

Comparison with the risk attributable to participating in traffic

A different approach is to compare the risk caused by the dose from the slag to the risk attributable to being a participant in traffic. Driving on a road there is a chance of dying in a car accident. The extra risk of dying attributable to the extra dose from slag received in the same time is a factor of about 1000 lower. However, these risks are not really comparable. Deaths caused by traffic are real and among them are relatively high numbers of young people. A dose caused by ionising radiation is thought to enhance the cancer occurrence in a given population. Consequently it is thought that there are relatively less young people involved compared to traffic deaths. Furthermore, there is no proof that these low doses will cause more cancers in a population. The risk value (extra chance of dying after being exposed to a dose or the number of deaths per man Sv), derived from the linear dose/response relationship hypotheses with no threshold value, is a number that actually only should be used to obtain safety standards for radiation protection. Consequently the deaths estimated with this risk value are hypothetical. This comparison only shows that the hypothetical risk is on a low significance level.

13.6 House building materials

The construction materials in the house-building sector in the Netherlands are subject to the standstill principle. Based on this principle, regulation is under development (Stralings Prestatie Norm: Radiation Performance Standard) with which the Dutch authorities can evaluate building designs in order to check whether the total dose caused by both radon emission through the crawling space and the use of construction materials in Dutch dwellings exceeds a specific threshold. In most cases the architect will have to use a program in which all relevant information of the specific dwelling and the radiological properties of the used materials have to be filled in. The result of the calculation can be compared with the threshold. In cases where the threshold is exceeded a redesign with the use of other construction materials will have to take place until the requirement is met. The individual dose caused by construction materials is in the range of 0 to 1000 _Sv per year for gamma-radiation. Radon in the Netherlands causes a dose in the range of 500 to 2000 _Sv per year of which the dose due to the emitted radon from the construction materials gives the larger part. It is possible that concrete produced with phosphorous slag could be used for piles in this system.

14 Position

Phosphorus slag is a building material, and should therefore not be subject to stricter standards than those applied to building materials in the house-building sector.

In this context it is clear that the release of slag only for road and hydraulic engineering applications already is the optimum to avoid unnecessary individual dose and collective dose. The caused individual dose and collective dose is a fraction of the variation in the natural background of which the phosphate ore initially was a part.

Abbreviations

^{238}U	Uranium with mass number 238
$^{238}U+$	Uranium with mass number 238 and daughters in equilibrium
ABC-factor	Actual exposure correction factor
AID	Actual Individual Dose
ALARA	As Low As Reasonably Achievable; social and economic facts taken into account.
AMAD	Activity Median Aerodynamic Diameter
Bq	Becquerel (the number of disintegration's per sec. due to RA-decay)
CE	Centrum for energiebesparing en schone technologie. A Dutch advisory organisation on energy and environmental issues.
Clearance level (conditional)	Activity levels above which disposal of radioactive matter is not allowed
COVRA	The Dutch central organisation for the storage of radioactive waste
dose	Effective dose: the sum of the equivalent doses, weighted by the appropriate tissue weighting factors, in all tissues and organs of the body
ECN	A Dutch Energy research institute
Exemption level	Activity levels above which a licence is necessary
HHNV	Hoechst Holland NV
ICRP	International Commission on Radiological Protection
KEMA	A Dutch research institute
Man Sv	Collective dose Man-Sievert
MID	Multi-functional Individual Dose
MID*	Multi-functional Individual Dose without the protection provided by housing
MTD	Maximum Tolerable Dose (40 or 100 _Sv per year)
NLG	Dutch guilders
PAS	Personal Air Sampling
Practice	Utilisation of radionuclides for their radioactive properties (functional)
RIVM	Dutch National Institute for Public Health and Environment
Sv	Sievert. The SI unit of equivalent dose. 1Sv=1 J/kg
TIBV	Thermphos International BV
VROM	The Dutch Ministry of Public Housing, Planning and the Environment
Work activity	Activity with enhanced levels radionuclides not processed for their radioactive properties (non-functional)