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Emissions of radionuclides by the Dutch phosphate industry. A review of the 'fluctuations' in the estimated risks

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EMISSIONS OF RADIONUCLIDES BY THE DUTCH PHOSPHATE INDUSTRY.
A REVIEW OF THE FLUCTUATIONS IN THE ESTIMATED RISKS.

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ABSTRACT

The radiological risks due to the emissions of the phosphate producing industry have been subject of discussion in the Netherlands for more than a decade. A survey of this discussion is presented. It is limited to the risks of the phosphogypsum effluents of the wet phosphorous plants in the Rijnmond area. The progressive changes in four important aspects of dose estimation are detailed for these industries:

1. Gradual changes in the phosphate production process and production capacity; a factor which is, however, of secondary importance compared to the next ones,
2. The growing scientific insight in the environmental behaviour of the various radionuclides released,
3. Policy changes with respect to the protection of man and its environment, and
4. Changing views of the metabolic behaviour and internal radiation dose of the radionuclides involved, resulting in changing dose conversion coefficients.

Changes in each of these factors induced major fluctuations in the estimated radiological consequences of the emissions. Successive results of dose calculations appear to range from hardly above the limit for a single source (100 mSv/a in the Netherlands) to far above this limit.
Firstly each factor will be treated separately. In conclusion a chronological series of a number of dose estimates will be given.

THE SOURCE

Two fertilizer industries, located along the Nieuwe Waterweg near Rotterdam, extract phophoric acid from phosphate ore by means of sulphuric acid. The waste product phopshogypsum is released into the river. Part of the radionuclides of the ^{238}U-series, which are present in activity concentrations of up to 2000 Bq/kg in some of the sedimentary phosphate ores [1], are released as well. Between 70% and 90% of the ^{238}U goes to the fertilizer, whereas about all ^{226}Ra, ^{210}Po and ^{210}Pb follow the gypsum [2, 3]. The two plants together released 660 GBq of ^{226}Ra in about 1.3 Mtonnes of gypsum in 1993 and 810 GBq in 1.4 Mtonnes in 1994. The average ^{226}Ra concentration of the effluent over these two years was about 550 Bq/kg [4]. The actually allowed emission for the two plants as a whole is almost 1000 GBq/a. The releases steadily decreased through the years. About 10 years ago, for example, more than 2 Mtonnes of gypsum were produced per year, with estimated levels of ^{226}Ra of more than 1000 Bq/kg [5, 6]. A progressive reduction of the emission was caused by a declining production and by the selection of other, less contaminated sedimentary ores or ores of magmatic origin (e.g. apatite with a ^{238}U-concentration of less than 100 Bq/kg) [1].

THE PATHWAYS

As the contaminants are released into the water, initial studies focussed on activity levels of fishery products. The transfer of ^{210}Po to shellfish was observed to be highest (20 m^3/kg) and that of ^{226}Ra to shrimp to be lowest (0.1 m^3/kg). These observations caused quite some effort to be spent on the study of the behaviour of polonium [7, 8].

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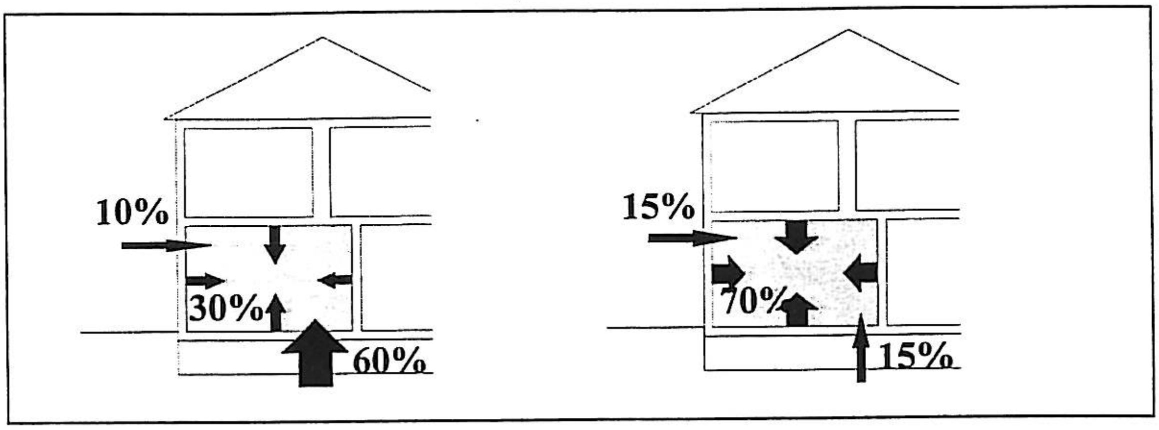

Figure 1 Relative contribution of the major sources of ^{222}Rn (building materials, crawl space and outdoor air) to the ^{222}Rn level of the average living room upon which all previous studies were based (left), and relative contribution determined in the national radon survey [15] (right).

When realizing that the contaminated dredged river and harbour sludges had been used for a long time in the Rijnmond area as landfill for building land [9], another exposure pathway came into the picture: inhalation of ^{222}Rn originating from the enhanced level of ^{226}Ra in the sludges. According to the first estimates this pathway was the most important one [10, 11, 12]. The rule of thumb was that each extra Bq/kg of ^{226}Ra results on the average in an enhancement of the indoor ^{222}Rn level with 0.8 Bq/m^3. This caused a major swing in the research on the emissions of the industries concerned. Extensive analyses of the contamination levels of the harbours and dumping areas were made and the importance of the soil as a source of indoor radon was studied in detail. Surplus concentrations of up to 280 Bq/kg of ^{226}Ra were found in the river [13] and of up to 120 Bq/kg on the dumping sites [14, 15], whereas the background was estimated to vary between 8 and 35 Bq/kg. The original estimate, however, of the contribution of ^{222}Rn originating from the soil to the ^{222}Rn level in the average dwelling had to be scaled down significantly as a result of the outcome of the 1995-1996 national survey on radon in dwellings (Figure 1) [16].

Other pathways such as the consumption of vegetables grown on contaminated dumping areas were considered of secondary importance [9].

THE EXPOSED

Up to now the individual risk or radiation dose has always been the one and only basis to assess the consequences of regular emissions of an industry. The exposed, i.e. the group of people for whom the level of exposure is estimated, were redefined occassionally. In former days the assessment was often based on a critical group composed of individuals with a superhuman appetite, living in severe conditions, and exposed via all potential pathways at the same time, even when their simultaneous occurrence could be excluded. In actual estimates of the individual radiation dose multifunctional use of the environment [17] still is one of the starting points [18]. Combinations of pathways should be realistic and the life-style of a member of the critical group in the Netherlands nowadays starts resembling that of the average man in the street [19]. A typical example of such a change is the pronounced adjustment of the consumption of fish (from 52 kg/a in [9] to 4 kg/a in [17]) and shellfish (from 10.4 kg/a in [9] to 0.3 kg/a in [17]), which made almost vanish the relevance of this exposure pathway.

Especially with respect to the definition of the exposed, the absence of a clear-cut border between the responsibilities of the scientist and those of the policymaker always has been a cause of major discussions [12].

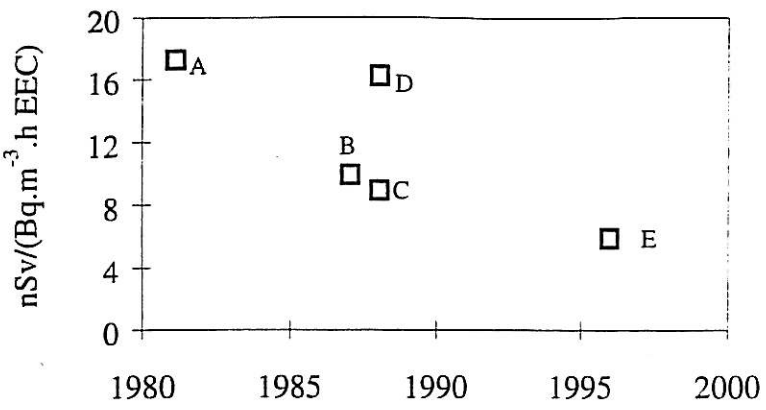

Figure 2 DCC for ^{222}Rn progeny in A: ICRP-32 [20], B: ICRP-50 [21], C: UNSCEAR [22], D: Nazaroff and Nero [23] and E: ICRP-65 [24].

THE DOSE

Changing insight in the metabolic behaviour of nuclides, which was translated into amended model calculations, and the regular (re-)analysis of epidemiologic data resulted in a 50% decrease of the dose conversion coefficient (DCC) for ^{222}Rn (Figure 2) over the past fifteen years. Over the same period the DCC for ^{210}Po increased sixfold, from 2.1£10^{-7} Sv/Bq [25] to 1.2£10^{-6} Sv/Bq [26].

REVIEW OF DOSE ESTIMATES

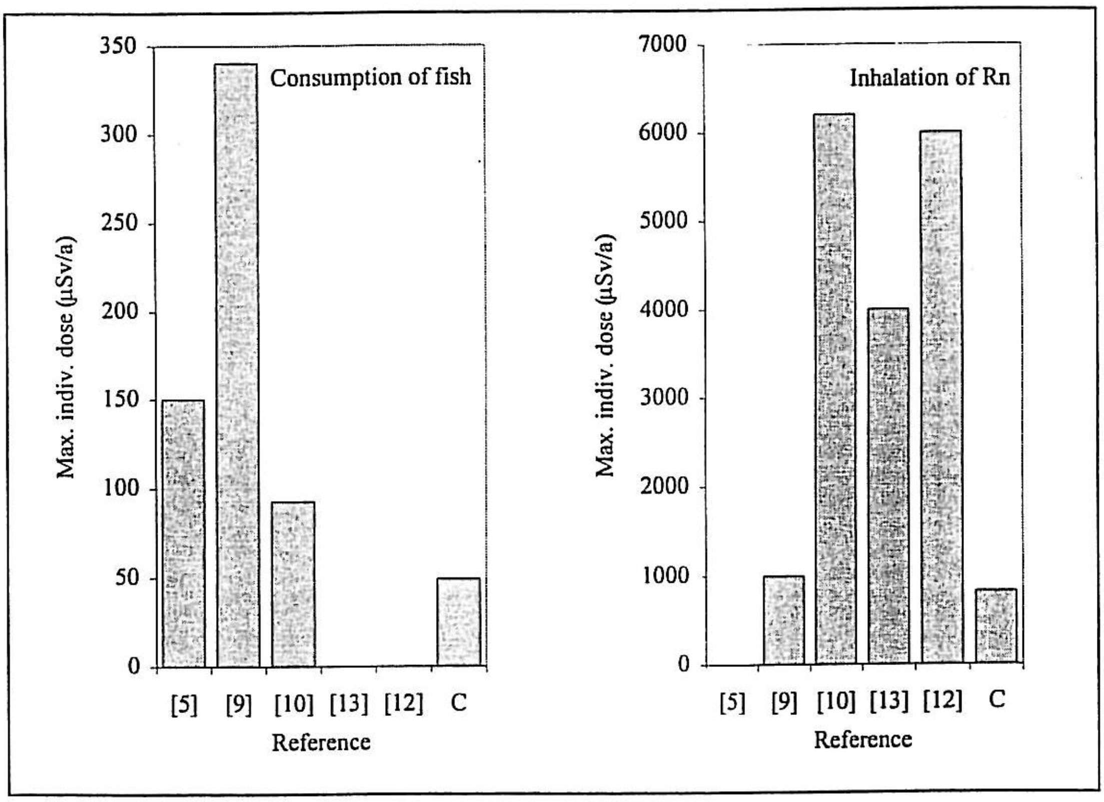

Figure 3 Estimates of the radiation dose via seafood and when living on a dumping site of harbour sludges in successive studies (C=current paper, [10]= sum of the estimates of the individual plants made in [10] and [11])

A chronological review of dose estimates (Figure 3) reveals major differences of up to a factor of 10 for the pathway consumption of fishery products and of 7.5 for inhalation of radon . Some of the differences in starting points or parameter values are hardly traceable and of minor importance. The bigger ones have been discussed before. For example: the results of Bos *et al.* [10, 11] are for extreme fish consumers , for the allowed emissions and based on old DCCs, whereas the current estimate is based on actual DCCs (dose estimate £ 2) [24] and emissions (dose – 2), and on normal fish consumption (dose – 3).

These data clearly demonstrate the need for:
1. A standard framework for the assessment and comparison of risks,
2. A standard framework for reporting on assessments, which clearly states starting points and the background of updates and deviations from prescribed procedures.

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