Environmental radioactivity: A perspective on industrial contributions

Marine studies are helping to document how some conventional "non-nuclear" industries enhance the natural radiation environment

by M.S. Baxter

Awareness of exposure to natural radioactivity — and its technological enhancement — in both the industrial and environmental domains is growing. Unfortunately, common errors frequently arise, namely associating ambient radioactivity only with the nuclear industry, and indeed the assumption that radioactivity is in some way unnatural.

The story is so basic that it begins at the birth of the universe. It was the combination of nuclear reactions and radioactivity that created all matter. It was nuclear stability which determined which elements are abundant and which are rare in our universe. Of the more than 5000 known kinds of atom (nuclides), about 95% are radioactive. It is the norm. Virtually all materials and environments on our planet are both radioactive and naturally exposed to ionizing radiation. The energy (heat) from this radiation has powered the major geological changes on earth, its division into core, mantle, and crust, its internal convection cycles and their exterior earth-shaping manifestations such as earthquakes, volcanic activity, mountain building, continental drift, and so on.

The natural radiation environment also has triggered and catalyzed some of the key stages in the very evolution of life. And, of course, the fusion reactors — our sun and the stars — have provided the nuclear energy which is the primary source of our daily light, our heat, our climate, and indeed our alternative and secondary energy sources such as coal, oil, gas, wood, peat, wind, and others. Radioactivity in rocks similarly feeds geothermal energy supplies. Thus, almost all industrial and domestic heating sources are primarily of nuclear origin and, as commented above, all natural materials are radioactive. Indeed, it can also be noted in passing that the natural radionuclides provide a unique means by which to learn about the rates and mechanisms of natural processes, since radioactive decay is Nature's only truly independent "time clock" having a known rate by which we can date rocks, sediments, archeological remains, and other materials.

Typical radioactivity concentrations in a range of everyday materials vary considerably. *(See table, page 35.)* For reference, it should be borne in mind that the lower activity limit for radioactive materials — that is, above which a substance is considered radioactive — has traditionally been set in the range 100 to 400 microsieverts per year for solids, depending on context and usage.

The fact that radioactivity levels in many everyday materials lie above these limits does not of course imply that they represent a significant hazard. The exposure route and duration must be taken into account in such an assessment. But the key point is that the naturally occurring radionuclides — primarily potassium-40 and those belonging to the decay series of uranium and thorium — occur at sufficiently enhanced concentrations in both natural and man-made materials that they *could* be regarded as radioactive substances and *could*, under certain circumstances, generate a significant radiation exposure.

The increased awareness of the importance of natural radioactivity as a source of general everyday radiation exposure is reflected in the changing estimates by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Its estimates of the average natural dose to a member of the public from the natural decay series radionuclides have changed

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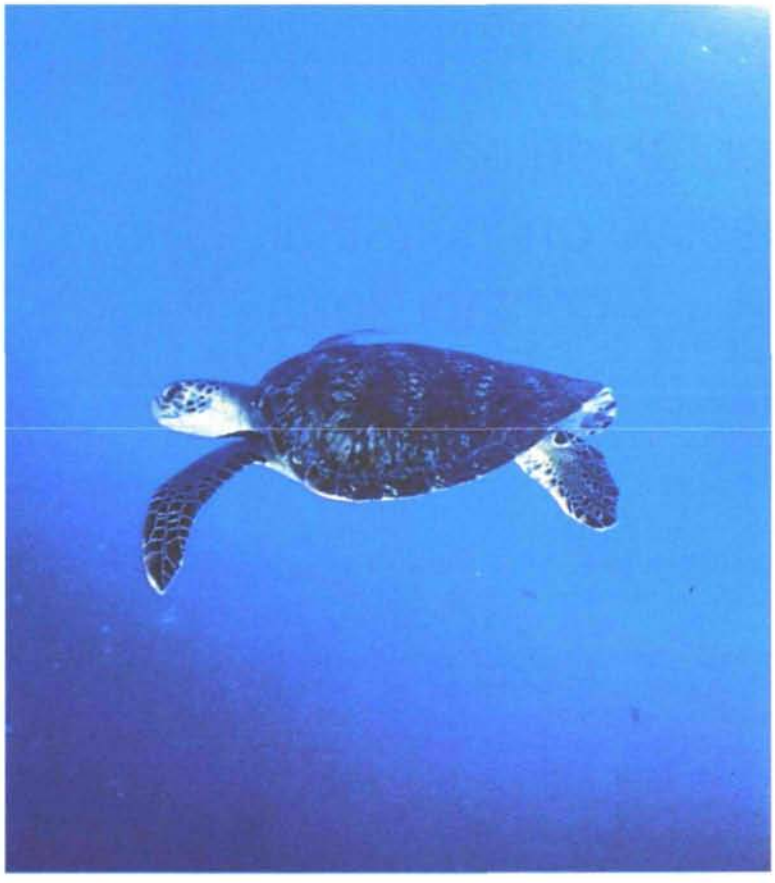
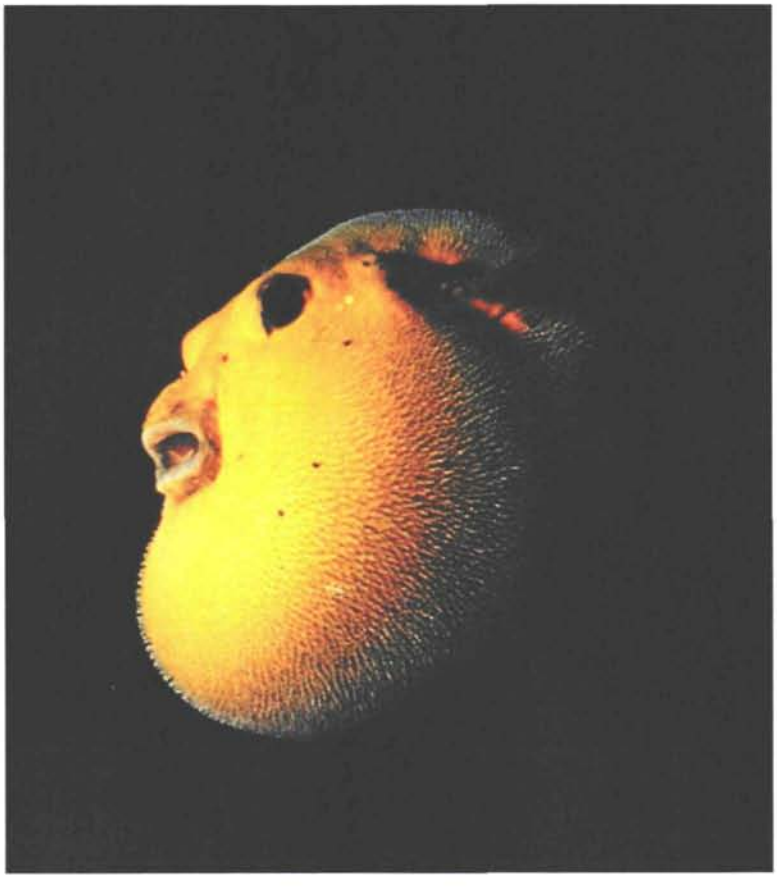




from about 50 microsieverts per year in 1962, to about 150 in 1972, to 1040 in 1977, to 1140 in 1982, and to about 1400 today.

This essay offers a perspective on the ubiquity of radioactivity in nature, one which builds upon the fundamental nature of nuclear processes in supporting our actual existence and emphasizes the significance of natural radioactivity to our everyday radiation exposure. Some "conventional" and "old-fashioned" industries are briefly cited that routinely use and have always used materials containing natural radioactivity and which concentrate and then release enhanced levels of these radionuclides to the environment.

The aim is simply to improve understanding of, and not to blame, such industries and perhaps to encourage a move towards a more objective and fairer comparative assessment and awareness of the environmental impacts of different industrial activities.

Radioactivity and fossil fuels

The best documented but perhaps still unappreciated input of enhanced natural radioactivity is from fossil fuels, namely coal, oil and gas. Their radioactivity results from the significant content of uranium, thorium, radium, radon, and polonium isotopes which are enhanced and then released during fuel extraction and burning. One recent review shows that, typically, the generation of one gigawatt-electric (GWe) of coal-fired electricity results in the environmental release in fly ash and off-gases of around 10^9 to 10^{11} becquerels (Bq) per year of both radon-220 and radon-222 and 10^8 to 10^{10} Bq per year each of lead-210, polonium-210, radium-226 and 228, thorium-232, and uranium-238.

These routine release rates, largely to the atmosphere, are less than or comparable to release rates from nuclear power stations under normal operation. They also globally add to the environment around 5000 tonnes of uranium, 8000 tonnes of thorium plus all their daughter products, including about 600 terabecquerels of alpha-emitters. The main result is a collective dose commitment of about 200 man-sieverts, with a typical critical group dose rate of up to 50 microsieverts per year.

The natural radioactivity in the fly ash waste product is concentrated by volatilization and sorption processes relative to the original coal burned. *(See table, page 37.)* One immediate consequence of this enrichment of radioactivity

Studies of the marine environment have been instrumental to scientific understanding of natural radioactivity and its exposure pathways. ***(Credit: Aldo Brando, Bogotá)***

Typical concentration (range)			Concentrations of principal nuclides			
Note All concentrations in becquerel per kilogram			Alpha			Beta/gamma
Material	**Alpha**	**Beta/gamma**	**Radon-226**	**Thorium-232**	**Uranium-238**	**Potassium-40**
Rocks						
Igneous	140	800	48	48	48	800
Granite	170		90	80		
Sedimentary sandstone	64	330	26	14	24	330
Sedimentary shales	95	800	40	40	15	800
Limestones	36	80	16	5	15	80
Phosphate ore	1500	260	1400	50		260
General soil	550 (300-1000)	440	70 (7-180)	40 (4-100)	24 (8-110)	440 (0.2-1200)
Bricks	110 (7-170)	600 (10-1000)	60 (2-90)	46 (3-80)		600 (10-1000)
Gypsums & cement	300 (30-800)	90 (40-160)	300 (20-800)	25 (8-60)		90 (40-160)
Sands & gravels	200-2000	30	4	20-200	20-90	30
Concretes	90 (40-170)	500 (210-650)	60 (7-140)	30 (13-42)		500 (210-560)
Human organs & tissues	0.2	67 (including rubidium-87)	0 003	0.0002	0.003	60
Books	30	100	0.9-30 (polonium-210)			100
Coal						
Yorks 1982	60	300	20	20	20	300
UK 1984	82	170	15	12	14	170
Fly ash	1400	1100	200 (600 polonium-210)	200	200	500
Fertilizer						
Normal superphosphate	2200		770	20	740	
Concentrated superphosphate	4600		800	10	2000	
Dried grass		600	300 (polonium-210)			600
Seaweed (Cornwall)	17				8	

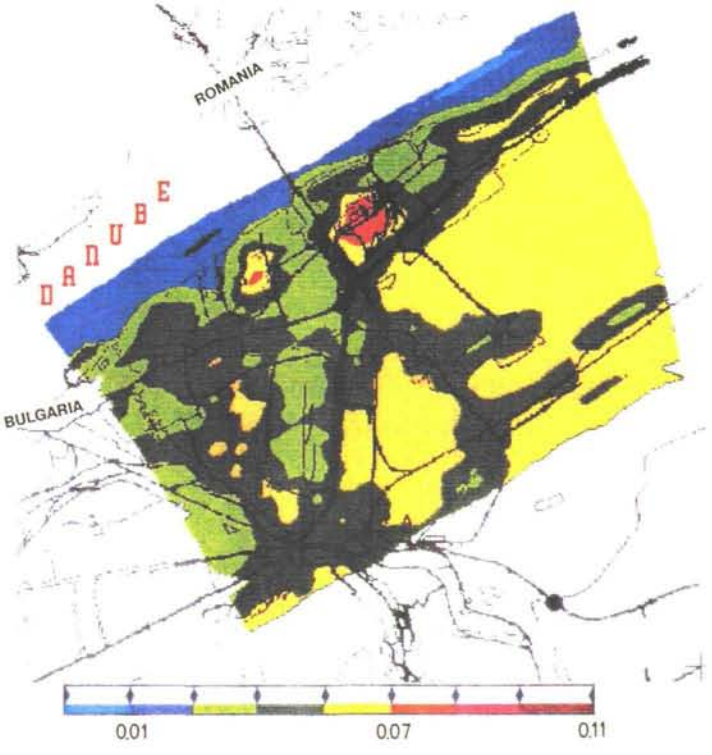
Levels of natural radioactivity in common materials

in fly ash is that its common use in the production of construction materials results in correspondingly higher gamma-dose and radon-exhalation rates in buildings and environments built from bricks and building blocks manufactured in this way.

The fly ash released in coal-burning is in the form of vitrified fine particles (1 to 100 micrometers in diameter). Despite stack filtration and other trapping methods, soils in the environment of coal-fired power plants are often found to be enriched in such ash-borne radionuclides. Ash collected by stack filters and coal residues also is often disposed of locally in dumps or wet lakes and these again show enhanced levels of radioactivity.

An example of such an increased radiation environment comes from a recent aerial gamma-spectrometric study of the Danube River Basin. In this project, scientists from the IAEA Marine Environment Laboratory (MEL) in Monaco cooperated with the French Commission of Atomic Energy (CEA), the Equipe Cousteau, and local Member State departments and institutes to survey the near-river environment of the Danube. They used a large-volume gamma-detector

Under an IAEA-MEL project, a helicopter-based survey was done in 1992 of typical fly ash disposed of into a lake near the Danube river in Bulgaria. The map shows gamma radiation dose rates for one of several industrial areas in which conventional industries were found to influence the local radiation environment significantly. The yellow areas near the Bulgarian city of Russe show enhanced gamma activity from potassium-40. These areas are observed to correlate well with the agricultural fields treated with potassium-rich fertilizers. Nearer the Danube itself (which is blue because of the low radioactivity content), two smaller orange/red areas show increased bismuth-214 signals in the airborne spectrometers, indicating the presence of its parent, radium-226. Subsequent investigation by scientists in Bulgaria showed that the "hot spots" result from partially filled-in sedimentation ponds into which the ash and mineral waste from a 200-MWe coal-fired power station are transported and then allowed to settle. The ponds drain into the river. Subsequent analyses at Sofia University and IAEA-MEL showed that the neighbouring soils contain 500-700 becquerels per kilogram (Bq/kg) of potassium-40, about 160 Bq/kg of radium-226, about 60 Bq/kg of thorium-232, about 50 Bq/kg of lead-210, and about 75 Bq/kg of polonium-210. The gamma dose rate is enhanced at the site by about 70%. The maximum space-averaged dose rate measured from the helicopter is about 100 nanogray per hour, which is at the high end of the normal environmental range. (A recently published radiation atlas by the Commission of the European Communities shows that typical outdoor gamma dose rates within Europe range from less than 30 to more than 80 nanograys per hour, with a mean of about 50 nanograys per hour). Although the ash and soils show enhanced radioactivity and the site itself is unfenced, there is not necessarily a radiological problem. The enhancements are rather mild and the dosimetry depends on local pathways and habits. The likely exposure pathways are through the water draining into the Danube and thence to the riverine food chain and by inhalation and ingestion of windblown dust rich in fly ash. In this particular context, however, the local hygiene inspectorate was informed so that a full assessment could be made. Such observations of enhanced radioactivity are not specific to Bulgaria: they are found in all countries and wherever coal is burned.



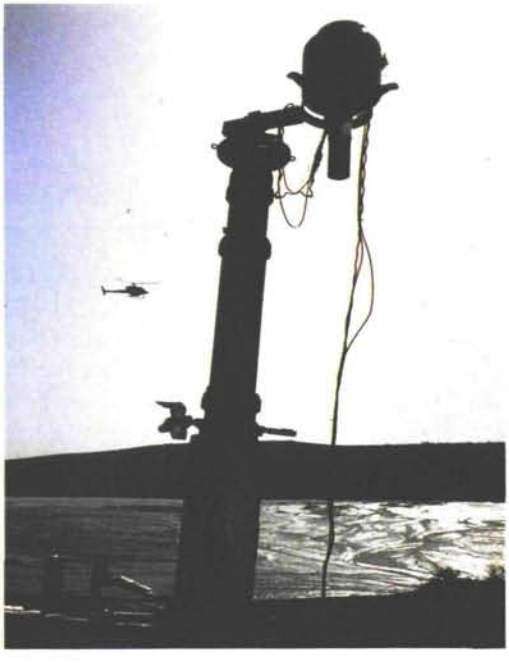
Note: Irradiation in microgray per hour.
Scale: 1 cm = 359 m

mounted below a helicopter, a most efficient and rapid means of mapping both natural and man-made radiation in the environment. *(See box, photo.)*

Oil and gas burning is characterized by similarly interesting interactions with the natural decay series nuclides, particularly, in these cases, with the radium and radon isotopes. Thus, for example, radon-222 diffuses into natural gas and oil deposits within the earth and it and/or its daughter nuclides are subsequently released in the power plants or dwellings where the fuels are burned. It has been estimated that the critical group near a gas-fired power plant can receive an effective dose equivalent of up to 20 microsieverts per year through ingestion of radon-222 progeny in seafoods and leafy vegetables. A similar dose contribution can result from radon-222 inhalation in homes burning natural gas.

Perhaps one of the better documented environmental exposure pathways in this area concerns the nuclide concentration process which occurs within brines raised to the surface during extraction of oil and gas. Salts from oversaturated brines — typically alkaline earth metal sulphates and carbonates — precipitate out and solidify as "scale" on the insides of pipes, pumps, or tanks. This eventually reduces and blocks the flow of fluid and necessitates dismantling and removal of the scale.

Because the main natural cations in these studies are calcium, strontium, and barium, they are extremely efficient in scavenging and concentrating their close relative radium, namely the isotopes radium-226 and 228, from the brines, groundwaters, and seawaters. Concentrations of



radium-226 and 228 up to and exceeding 10^6 becquerels per kilogram (Bq/kg) have been observed. Workers who clean out and decontaminate such equipment must follow strict safety procedures comparable to those in high-activity nuclear laboratories.

It is not uncommon, however, for the effluents from industrial sites of this kind to discharge directly to the environment. We have, for example, recently noted enhanced polonium-210 concentrations (625 Bq/kg) in marine organisms collected in the vicinity of such a pipeline discharge. Incidentally, the same study, conducted by the Scottish Universities Research and Reactor Centre, also observed enhanced concentrations of lead-210, polonium-210, thorium-232, and uranium-238 in coastal sediments collected near a site where coal mining waste is discharged directly into the sea.

It must not be concluded that the radioactivity released by the fossil fuel power industry is either extensive or harmful. However, in some cases, the releases can be significant on local scales and are considerably less understood and controlled than those of similar magnitudes by the competing nuclear industry.

The fossil fuel industries already know the problems of global climate change, acid rain, pollution by toxic metals and organic compounds, mining accidents and explosions during distribution and use, plus the undesirability of burning a valuable and finite natural resource from which, for example, pharmaceuticals and polymer materials can be manufactured. Greater understanding and consideration of the environmental radioactivity enhancements resulting from the use of fossil fuel certainly should provide a further parameter for political and scientific decision-making.

Other conventional industries

The IAEA-MEL in Monaco, particularly through the work of Prof. Robin Cherry and co-workers, has been at the forefront of research showing that in nature marine organisms concentrate polonium-210 from seawater. They thereby receive a considerable local dose to individual tissues, notably to the hepatopancreas in which concentrations of up to several kilobecquerels (kBq) per kilogram (wet) result in doses from polonium-210 alone in the 100 millisieverts per year region. This marine organ therefore appears to experience one of the highest, if not the highest, naturally occurring radiation dose.

However, it was not until the mid-1980s that Dr Scott Fowler of IAEA-MEL and myself, with the help of a joint research student (now Dr Paul McDonald), showed that, even in the vicinity of a major nuclear discharge (near Sellafield in the United Kingdom), the natural polonium-210 alpha radioactivity of common edible mussels exceeded the alpha activity of the critical group exposed to artificial radionuclides. Polonium-210 activities ranged from 124 Bq/kg in muscle to 600 Bq/kg in viscera.

In a follow-up study, Dr McDonald, myself, and co-workers then showed that within the Irish Sea coastal zone there indeed was an additional and rather large source of polonium-210 from the non-nuclear industry. Concentrations of 0.3 to more than 3 kBq/kg of polonium-210 were observed in mussels from the Whitehaven region of the UK. These enhancements resulted from

Natural levels of radioactivity in selected materials

Total concentration (alpha + beta/gamma)	
From 100 to 400 becquerels per kilogram	**More than 400 becquerels per kilogram**
Cereals	Tea
Meat	Coffee
Poultry	Dried mushrooms
Potatoes	Some shellfish
Some green vegetables	Some Brazil nuts
Root vegetables	Some drinking water
Some fresh fruit	Some rocks
Fruit products	Soils
Some beans	Some bricks
Fish, some shellfish	Some gypsums
Some Brazil & other nuts	Some concretes
Some drinking water	Fly ash
Some rocks, bricks	Fertilizers
Some gypsums	
Some concretes	
Cement, sand, gravel	
Books	
Coal	

Comparative levels of radioactivity in fly ash and coal

Activity content in becquerels per kilogram								
	Potassium-40	**Uranium-235**	**Radon-226**	**Lead-210**	**Polonium-210**	**Thorium-232**	**Thorium-238**	**Radium-238**
Coal	50	20	20	20	20	20	20	20
Fly ash	265	200	240	930	1700	70	110	130

routine releases of wastes from a phosphate processing plant (now closed).

A follow-up study by the UK Ministry of Agriculture, Food, and Fisheries showed that the wastes from this detergent manufacturing factory also contained significant inventories of radium-226 and thorium isotopes. Critical group doses to local seafood eaters were in the 0.3 to 3 millisieverts per year range, in excess of any value resulting at that time from nuclear discharges in the UK.

Similar environmental enhancements of natural decay series nuclides have been observed quite regularly in the vicinity of phosphate processing plants, reflecting the natural chemical affinity of uranium and radium for phosphate ion and their accumulation in phosphogypsum waste products. The fertilizer and detergent industries are the main focuses for the release of enhanced natural radioactivity in phosphatic wastes.

One of the best known examples of this phenomenon is the discharge of phosphogypsum effluents from the industrial zone at Rotterdam in the Netherlands. Dr Heko Köster of the National Institute of Public Health and Environmental Protection in the Netherlands and his co-workers have shown that about 10^{12} Bq per year each of polonium-210 and radium-226 are released. They lead to enhancements of about 100 Bq/kg of polonium-210 in edible parts of mussels and prawns living 50 to 100 kilometers from the industrial zone. Individual dose rates of 0.1 to 0.3 millisieverts per year are predicted amongst groups of seafood consumers, while the use of contaminated harbour sludge as landfill in polders (reclaimed land) around Rotterdam can generate individual doses of 0.3 to 1 millisieverts per year from consumption of local livestock products and from inhalation of enhanced indoor radon-222.

This phosphate-centred example typifies a rather large number of other "conventional" industries which, mainly because their feed materials are rich in natural radioactivity, can and do release enhanced radioactivity to the environment.

I was recently requested to review the radioactivity flows into, through, and outside one of the largest ore smelters (tin, copper, lead, etc.) in the world. With a small team of helpers, we discovered that feed materials had polonium-210 contents up to 66 kBq/kg and that industrial processing further concentrated the radionuclides. For example, intermediates had polonium-210 concentrations of up to 2.2 megabecquerels per kilogram. The annual flow through the factory was in the terabecquerels per year range and, besides stack discharges, the wind-blown dusts of contaminated waste provided a potential environmental dispersion pathway.

Similar radioactivity enhancements are associated with industries such as production of titanium oxides, rare earth compounds, mineral waters, paint and ceramics, and in the use of tailings from the alum shale industry and of zirconium-rich sands — that is, in the colouring, clothing, lime burning, oil, and construction industries. In all these cases, natural decay series nuclides occur at relatively high concentration in the source materials and can be enriched and discharged by the industrial process.

The list could continue. But the message should end here. That message starts with the fact that "conventional industries" often generate environmental radioactivity enhancements. As mentioned earlier, we have recently even reached the stage where, in at least one major "nuclear" country, the United Kingdom, the potential maximum dose rate to a critical group of the public has been higher from a "non-nuclear" industry (phosphate-processing) than from one of the largest nuclear sites in the world (Sellafield). Like the radioactivity exposures induced by the nuclear industry, few if any of these enhancements by "conventional" industries are really significant in the health context either on global or local scales.

A move toward greater balance

We live in, and have to be thankful for, a radioactive world, with past and present variabilities of the natural background radiation field which far exceed the trivial man-made effects discussed here. What is significant when comparing the nuclear and non-nuclear industries is the imbalance in (1) the understanding and control of, (2) the required safety standards associated with, and (3) the financial responsibilities for minimizing these radioactivity emissions.

There is now an international move towards harmonizing the assessment and control of both sets of nuclide sources, that is to bring the emission control standards of the non-nuclear industries into line with those more stringently applied within the nuclear industry. Once the radioactivity is treated more equally both on the basis of impact assessments and costs, then the pressure similarly should be on to quantify the environmental and health detriments of the non-nuclear pollutants discharged from "conventional" industries.

It will, however, be some time before the exposure-effect relationships for non-nuclear industrial contaminants are as well defined and then controlled as the radioactive releases assessed by — but not restricted to — the nuclear industry. ❐