Assessing the radiation risk to workers exposed to thorium

In the context of actual or suspected situations of exposure to ionizing radiation, requests for health risk assessments have been made of the public experts in the field, OPRI and/or IPSN, that were merged to form IRSN in 2002. Audits were carried out jointly by OPRI and IPSN at Pargny-sur-Saulx in response to the demand made by the Prefect of the Champagne-Ardenness Region. In 1997 drums of radioactive waste were found on the site of Orflam-Plast, a company now in liquidation, revealing the past exposure of its former workforce that had been unknown and whose severity had to be assessed.

Handling monazite was brought to a halt in the last phase of the company as the lighters were assembled from purchased components. So in principle two different risk periods relating to thorium can be distinguished for the site employees and the local population:
• in an early period (1934-1970), the potential exposure and contamination risk was severe because the thorium ore was being handled;
• from the 1970s onwards, the risk of external exposure was greater than that of internal contamination on the Orflam-Plast site, although the risk presented by the presence of thorium in the dike leaching waters is an unknown quantity. It was thus essential that a radiological risk assessment be made.

IRSN carried out a series of surveys from 1997 until 2001 for the purpose:
• geographical study of mortality;
• control case studies;
• radiotoxicological analyses.

Geographical study of mortality

An audit carried out by IRSN in 1997, demonstrated that the former employees and neighboring population were likely to have been exposed to ionizing radiation at levels considerably higher than naturally occurring radioactivity levels, the dose rate occasionally peaking at 10 μSv/h, that is over 100 times the natural ambient level. These estimates, based on the measurements made in the defunct manufacturing environment, did not enable any conclusions to be drawn on earlier potential exposure. Moreover it appeared that the majority of

The records of the former Orflam-Plast manufacturing plant at Pargny-sur-Saulx, revealed that from 1934 to the beginning of the 1970s, significant amounts of imported monazite ore, rich in natural radioactive elements, particularly thorium-232, with a half-life of 14 billion years had been handled. At that time, Orflam-Plast chemically extracted the ore, cerium, a hard metal used for making lighter flints. The residual waste of this manufacturing process was in solid form made up of mineral sand with high thorium content. The several thousand metric tons of monazite handled in this plant thus led to an initial approximation of an almost equivalent amount of waste. Apparently no precautions were taken when spreading this ore residue whose radiological content was ignored or unknown in the environment. It was found scattered on the very site of the company and in particular, in the premises where the ore was handled in bulk. About 7500 metric tons of this residue was used to build an embankment several hundred meters along the River Saulx, both upstream and downstream of where it crossed the plant site; the residue was thus “put to use” protecting the neighboring meadows from winter floods.

References
Occupational exposure to ionizing radiation was cited as potentially responsible for the surplus of cancers observed.

Orflam-Plast employees likely to have been exposed the most were recruited and lived in the districts surrounding Pargny. This led to a study into cancer mortality in conjunction with INSERM’s U521 unit from data from the national mortality database, managed by INSERM’s joint service 8 (SC8) at Le Vésinet. This study was carried out across all the districts within a 16 km radius of Pargny, for the period 1968-1994. It revealed an overshoot of 40% over the norm of deaths through lung cancer and bladder cancer in men aged over 40 living in the catchment area at the time of death. Moreover, this mortality surplus increased significantly as the study approached the district of Pargny-sur-Saulx (11 additional cases of lung cancer and 6 additional cases of bladder cancer).

A precise mortality analysis did not reveal a surplus of deaths through cancer in the neighboring districts of the Saulx downstream of Pargny. This observation enabled us to rule out the risk of a major health implication that might have resulted from thorium contamination carried by leaching waters. Consequently, the significantly higher than average national mortality rate through lung and bladder cancer registered in male residents of Pargny prompted the search for a one-off possibly occupational origin. This is why occupational exposure to ionizing radiation was cited as potentially responsible for the surplus of cancers observed, and in particular lung cancers, because of the presence of thorium in the dust generated while handling the ore.

Control case study

It was necessary to test the hypothesis according to which death through lung or bladder cancer within a radius of 10 km of Pargny-sur-Saulx was associated with having worked on the Orflam-Plast site in order to go a step forward. This epidemiological study of control cases of cancer mortality at Pargny-sur-Saulx was conducted in conjunction with the Environmental Health Department and the Health Watchdog Institute (InVS) and with the agreement of the French civil liberties organization, the CNIL. The individual data on death consolidated for the period 1968-1996 was supplied by INSERM’s SC8, for people living within a 10 km radius of Pargny-sur-Saulx at the time of death. For this control case mortality study carried out on the general population, the immediate, principal and combined causes of each death were obtained. The cases were defined as being individuals deceased through lung or bladder cancer in the study area and for the period under consideration. The control cases were defined as being individuals who had died from some other cause in the area under study for the study period. They were selected from a file of causes of death excluding respiratory ailments and genito-urinary apparatus ailments. Secondly these control cases were paired up to the cases by sex, age (in 5-year segments) and the period of occurrence of death. Three such periods of occurrence of death were...
Protection of human life

Orflam-Plast site (March 1999).

defined, in principle, on the basis of the mortality survey findings for the general population: period No. 1, 1968-1977; period No. 2, 1978-1987; period No. 3, 1988-1994. A fourth period (1995-1996) was added to draw on the maximum possible number of cases. Three paired control cases were associated with each of the other cases by random selection.

The characterization of exposure to thorium was carried out as follows. Research was made to find out for all the cases and control cases selected, whether they had or had not worked at the Orflam-Plast plant, however long the period of employment. This research was based on the register of former Orflam-Plast employees, held by the Champagne-Ardennes Regional Employment, Work and Professional Training Office (DRTEFP) in the occupational medical inspection department. The file was compiled from occupational medical records (for employees who worked on the site from 1980 onwards), the work inspection register (register of employees hired and leavers) and the register of employees covered by redundancy schemes. For each real and control case, a search was made on the "date of birth" identifier cross-referencing the file of medical causes of death against the file of former employees. As this common identifier could be insufficient to confirm whether the same individual was pinpointed, it was necessary to check for consistency between names and dates of death in the official registers. Of the 692 individuals listed in the former Orflam-Plast employees' file, the birth date details of 46 were unknown. INSERM's unit 472 was requested to supply the life statistics of these people and find their dates of birth, in application of decree 98-37 "authorizing access to data relating to the death of people registered in the RNIPP in the context of research into health". Eleven of those 46 individuals were subsequently identified.

The statistical analysis of the data consisted in comparing the frequency of membership of the category of former Orflam-Plast employee in the real and control cases by making a paired analysis (Epi-info software, version 6.04).

**The Findings**

The mortality data for the study between 1968 and 1996 are as follows: 3501 deaths of which 879 cancers, broken down into 154 cases of lung cancer (142 men and 12 women) and 39 cases of bladder cancers (27 men and 12 women). The file of former employees of the manufacturing site numbered 692 individuals. The proportion of female employees was 74%. Of the 641 employees for whom information was available, 380 (59%) were hired before 1970. Of the 599 employees for whom information was available, 434 (72%) were aged under 20 when they were hired to work on the site.

- As regards cases of lung cancer, only 3 of the 150 cases to which 3 control cases could be attached had worked at Orflam-Plast, whereas 7 of the 450 corresponding control cases were former workers on the site. The paired odds ratio (OR) (statistical parameter measuring the excess risk) was 1.29 (confidence interval at 95% calculated according to the exact Fischer method: 0.27–4.89).

As the confidence interval is either side of 1 for which the risk is zero, there is thus no significant statistical association between the fact of death through lung cancer and having worked at Orflam-Plast.
• There was no case of bladder cancer among the former employees and only one in the control group of 117 individuals. As only 1 of the 39 groups did not conform the OR could not be estimated. Thus the control case study does not enable us to explain the higher mortality rate through lung or bladder cancer observed in men in the geographical mortality study. Furthermore in view of the fact that the expected number of lung cancers in the essentially female worker cohort, is low – around 3 – it is unrealistic to suggest a cohort study limited to the factory workers: effectively it would carry very little weight and would only detect very high excess mortality, a factor of 3 or 4, which on the basis of the state of the art, is very unlikely.

Radiotoxicity studies of the urine of former workers

The high lung and bladder cancer figures revealed during the geographical mortality study prompted the immediate implementation of thorium contamination studies without waiting for the results of the control case study. Thorium elimination from the human body is effectively very slow because this element is retained for a long time in certain organs, in particular the lungs and liver.

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If the results observed in these 10 former employees were negative, the probability of observing a positive result in another, according to the survey less exposed employee in principle, would be considered as zero. If the opposite were to apply the study should be pursued. The urine of these 10 employees was collected for 24 hours in two-liter bottles with sulfamic acid as a preserving agent. The radiotoxicity analysis of the urine comprised the following radiochemical and count stages:
• actinide coprecipitation – and thus thorium – in an ammonium medium by calcium phosphates and magnesium phosphates;
• following concentrated nitric acid treatment, the thorium $^{4+}$Th is trapped on an anionic resin, then eluated by concentrated hydrochloric acid;
• the thorium fraction not bound to the anionic resin is specifically fixed on TEVA resin in a nitric medium, then eluated by concentrated hydrochloric acid;
• the two eluates are put together and the thorium is coprecipitated by lanthanum fluoride;
• the isotopic half-life and identification of the thorium are established by making an alpha-spectrometry count of the precipitate.

The addition of a tracer, 20 mBq of $^{229}$Th, to the initial urine prior to the radiochemical preparation stages, makes quantitative analyses of $^{232}$Th and $^{228}$Th possible.

This methodology is very time-consuming: it takes two full weeks of work to conduct the three analyses. Interpreting the results is a complicated matter as it has to take into account the presence of varying quantities of natural $^{228}$Th and $^{232}$Th in the urine (approximately 300 µBq, $^{1}$ on average). The results show that 2 of the former employees have higher than normal readings for natural thorium elimination.

These preliminary findings thus dictated that a wider study be made using an ICP-MS, the only mass spectrometry technique today likely to improve radiotoxicity analysis protocols in terms of sensitivity and response speed. Compared with alpha-spectrometry, the use of ICP-MS considerably reduces the measuring time for long-lived radioelements (several minutes instead of several days for concentration levels of about 1 mBq, $^{1}$).

The first work carried out involved the study of a simple procedure for measuring uranium in urine. The results led to the proposal and validation of an ICP-MS uranium analysis protocol following simple dilution of urine by a factor of 20. Using this protocol, the detection limit, for example in natural uranium for a given apparatus sensitivity is 5.5 ng. $^{1}$ or 0.14 mBq. $^{1}$.

In the context of the health monitoring of former Orflam-Plast employees, the LEMDI (IRSN Internal Dose Assessment and Modeling Laboratory) transposed the protocol validated for measuring uranium to measuring thorium in the urine. ICP-MS can only measure thorium isotope-232 as the half-lives of its other isotopes are too short. The protocol was perfected on the urine of adults not exposed to thorium surcharged with $^{232}$Th. A sampling campaign of urine over 24 hours was then carried out by IRSN at the end of June 2000 from former Orflam-Plast employees living in the three departments close to Pargny-sur-Saulx – the Marne, Haute-Marne and the Meuse. Only 79 workers out of the 110 contacted were willing to take part and collected their urine for 24 hours in the two-liter bottles provided. Sample preservation was provided by sulfamic acid and cold temperature.

The $^{232}$Th measurements were taken after diluting in a factor of 20 aliquot parts in 20 ml of urine in ultra pure nitric acid at 2%, a favorable medium for ICP-MS measurements. The standards were prepared from urine blanks to get over the matrix effects. The results were expressed in mass (µg/24 hr) and half-life (mBq/24 hr) in $^{232}$Th. Sensitivity in ICP-MS depends on the apparatus adjustments with each measuring campaign. Thus the detection limits obtained on the 79 samples vary from 0.48–2.7 ng/24 hr, or 2–11.1 µBq/24 hr.

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References
The results for the former Orflam-Plast employees show that significant values, i.e. situated above background level, are only observed in 53% of the cases. It should be noted that the 2 former employees whose results were higher than normal when measured by conventional radiotoxicity measurements did not take part in this voluntary campaign.

The results of the former Orflam-Plast employees were compared with those of 539 workers not exposed to thorium in the Hurtgen study. The maximum value of the series of measurements made on the Orflam-Plast site (0.10 mBq/24 hr) was of the same magnitude as the minimum value observed in Belgium by Hurtgen in an unexposed population (0.15 mBq/24 hr). We note that individual excretion of natural thorium through the urine varies widely with the physiological characteristics of the individuals and the contamination levels of food ingested. Consequently the variations observed in the results may be due to a difference in nutritional ingestion of thorium, which can be considerable within the population, particularly through mineral water.

It can be concluded that the elimination of thorium in the urine by the 79 former Orflam-Plast employees examined seems normal. To confirm this result, out of some fifteen urine samples representing those with the highest Th readings, the 228Th was measured by spectrometry – after chemical purification. These results enable the existence of significant contamination of these employees by thorium to be ruled out.

Reference

Conclusion

To sum up, the various studies carried out by IRSN did not enable:
• any statistically significant association to be revealed between having worked in the Orflam-Plast plant and having died from lung or bladder cancer within a 10 km radius of Pargny-sur-Saulx;
• any significant elimination of $^{232}$Th through the urine of former Orflam-Plast employees to be demonstrated.
Thus the results of these studies are inconclusive with regard to the high mortality rate through lung or bladder cancer observed in men included in the geographical mortality study. Moreover it enables us to rule out the responsibility for contamination by thorium as the cause of the high rate of cancer observed.
As regards any continuation of these studies, and more particularly the search for an explanation into the excess mortality revealed by the geographical study, there are two lines of investigation open:
• the first would certainly be to study whether the mortality excess can be explained by the socio-professional characteristics of the local population;
• it would also be useful to study the relation between death through lung and bladder cancer within a 10 km radius of Pargny-sur-Saulx and having worked in another industry in the district (for example, the brickworks). But that is quite another story.
This research could be envisaged if the local authorities were to formally request them.

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