## **REMEDIATION OF TPP PLOMIN SLAG/ASH TENORM LANDFILL IN CROATIA**

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# ABSTRACT

TPP Plomin is a 125 MW (thermal) coal powered plant. Built on the Adriatic coast, at the end of the Plomin bay in Istria region of Croatia, it started operation in 1970 using coal from local mines from the nearby Rasa area. Recently a new 210 MW (thermal) block has been added. The coal that had been mined in Istria is characterized by: (a) high sulfur content and (b) increased radioactivity which varies not only from mine to mine but also from one coal layer to another. Uranium-238 and radium-226 specific activities were measured regularly, extending from 25-4,900 Bq/kg and 28-2,000 Bq/kg respectively. Slag/ash generated by TPP Plomin is TENORM. With the start of TPP Plomin operation slag/ash landfill was set up in a small valley close to the power plant site as a heap of waste eventually covering an area of 24 ha. No engineering structures were built to contain the material. Due to variability in radioactive content of domestic coal, "dilution" of this coal with coals of low radioactivity throughout the pile is pronouncedly uneven, varying considerably both horizontally and vertically.

Because of its elevated radioactivity, the landfill has been perceived by the general public in Istria region as a growing environmental threat. A preliminary safety assessment was performed to estimate occupational exposures, public exposures and environmental impacts. Potential dose of 0.6 mSv/y was calculated for the "landfill supervisor" scenario, and 60  $\mu$ Sv/y for the "nearby resident" scenario. In absence of national regulatory guidance ICRP-60 and BSS were used. Remediation of landfill has been initiated, prior to the expected national regulatory developments aimed at TENORM management. Project is conceived as a prompt but partial solution. The choice to remediate promptly, as well as the decision on appropriate level of the present remediation, was motivated by the prospective benefits in terms of potential doses reduction for those who work on the landfill or on the nearby land, and by the cost of remediation. In addition to radiological consideration, the decision to remediate was also influenced by the public concern and anxiety.

### **INTRODUCTION**

### **Elevated radioactivity in TPP Plomin landfill**

TPP Plomin is a 125 MW (thermal) coal powered plant. Built on the Adriatic coast, at the end of the Plomin bay in Istria region of Croatia, it started operation in 1970 using coal from local mines from the nearby Rasa area. Recently a new 210 MW (thermal) block has been added.

The coal for the TPP Plomin operation, that had been mined in Istria for decades, is characterized by some typical features of coals lying in karst regions. In particular, it has: (a)

high sulfur content and (b) increased radioactivity which varies not only from mine to mine but also from one coal layer to another.

It was established that the coal contains 10-14% of sulfur and varying specific activities of radioactivity in the local coal were systematically monitored. It was found that uranium-238 specific activity varied from 25-4,900 Bq/kg with an average of 310 Bq/kg (1,2). Measured radium-226 specific activity was also elevated from 28-2,000 Bq/kg, with an average of 260 Bq/kg. Measured values of specific activities for potasium-40 and thorium-232 were not significantly greater then world mean values for coals.

As expected, the radioactivity of slag/ash was increased also. Collected flying ash had uranium-238 specific activity varying over years from 500-8,600 Bq/kg, and an average radium-226 specific activity of 2,600 Bq/kg (3). Bottom ash was having uranium-238 specific activity from 400-1,800 Bq/kg, and radium-226 specific activity of 800 Bq/kg. Slag/ash generated by TPP Plomin is TENORM (technically enhanced naturally occurring radioactive material).

During the last ten years the local coal was "diluted" with imported coals having low sulfur content and low radioactivity. Finally the local mines were closed and since then only imported coals have been used.

With the start of TPP Plomin operation slag/ash landfill was set up in a small valley close to the power plant site as a heap of waste generated by routine operation. No engineering structures were built to contain the material. The landfill is situated at the eastern coast of the Istria peninsula, approximately 5 km from the seaside, and some 10 km north-west from the urban area but with some of the houses less 1,000 m from it. The landfill is placed over the downstream section of the Bizac torrent, along the contact zone of proluvial fan of the Boljuncica river (at the non-inundated section of the Plomin bay), and the slope of karst hilly area surrounding the bay. Bottom of the repository is composed of nummulite limestone. Thus, a typical karst hydrology is present at the area where locally occurring groundwater flows directly towards the sea.

Slag/ash to be disposed of is transported with conveyers. Disposed material is leveled by machinery. The maximum thickness of the accumulated slag/ash does not exceed 8 m, whilst the present volume of disposed material is some 900.000  $\text{m}^3$ , covering an irregular area of 24 ha. Approximately 70% of landfill area is covered by a simple soil layer thick up to 60 cm and partially topped with grass.

In order to decreases erosional, derosional and proluvial processes, the landfill is provided with drainage system and a retention pond which diminishes transport and deposition of eroded terrestrial material (primarily disposed slag/ash) into the Plomin bay.

Mediterranean climate prevails, locally characterized by an average annual rainfall of 1,294 mm and occasional strong winds up to 11 m/s. Hence, aeollian transportation of disposed and uncovered slag/ash is quite possible.

For practical reasons the same landfill will be used for slag/ash generated by the operation of the new 210 MW block.

Croatia has inherited Yugoslav regulations on nuclear safety, although it presently has no nuclear programme and there is no nuclear material used in the country. But TENORM is neither defined by Croatian low, nor is it regulated. General regulations on radiation protection do not provide clear guidance on how to address the problem of TPP Plomin landfill.

#### The landfill and the environment

The landfill and its likely impact on the environment were monitored by both *in situ* and laboratory measurements.

Due to variability in radioactive content of domestic coal, "dilution" of this coal with coals of low radioactivity and random placement of dumped waste material at the landfill, the distribution of radioactivity throughout the pile is pronouncedly uneven, varying considerably both horizontally and vertically.

In situ gamma-spectrometric measurements were carried out using HPGe Ortec detector (resolution 1.74 KeV on 1.33 MeV cobalt-60, relative efficiency 21.6%) at several locations within the operating (and uncovered) parts of the landfill. Covered parts of the landfill as well as a location at the seaside were also included into the measurements. Measuring time was 1,000 sec. Samples were collected, all liquid samples were radio-chemically separated, and radium-226 was finally determined by alpha-spectrometric measurements using silicon charged particle Si(Li) surface barrier detector. The counting time for each measurement was 80,000 sec. Presence of natural radionuclides of uranium and thorium decay series as well as potassium-40. have been detected. Corresponding contribution of measured radionuclides to the absorbed dose rate was calculated for each of the locations (4). The results of measurement indicated, as expected, considerable variations of the absorbed dose rates (operating part of the landfill 280-350 nGy/h; covered part of the landfill 190-215 nGy/h; seaside 3,000 m from the landfill 110 nGy/h). Comparing contributions of uranium and thorium decay series to the measured doses on the operational part of the landfill and the covered one, a difference is noted. Contribution of thorium decay series is roughly equal for both of the areas (P(t)>0.05), while radionuclides from uranium decay series are significantly more contributing to the absorbed dose measured on the operational part of the landfill (at level of P(t)<0.01). Potassium-40 is contributing more to the doses measured on locations over the covered parts of the landfill (P(t) < 0.01). This may be ascribed to the presence of vegetation.

It is of particular interest to establish how well the landfill presently contains the deposited material.

Recently measured sediment samples and seawater from the Plomin bay did not reveal any significant influence of the landfill. Values obtained by radiometric measurements of sediments are in accordance with the previously obtained characteristics for the majority of Istrian soils: specific activity of uranium-238 in the bay sediments (65 samples from different depths) was found to be 21 Bq/kg and for radium-226 it was 19 Bq/kg. Specific activity measured in the catchment area of the Boljuncica river and in Istria's soils of uranium-238 is varying from 8-63 Bq/kg with an average of 24 Bq/kg; for radium-226 it was measured to be from 15-65, on average 24 Bq/kg (5,6,7). This supports the view that most of the material at the bottom of the

Plomin bay was brought by the Boljuncica river from its drainage basin and not from the TPP Plomin landfill.

It should be noted that on another site in Croatia (Kastela near Split, in Dalmatia region), a similar but considerably smaller pile of the same origin (burning coals from the mines in karst region) containing some 10,000  $\text{m}^3$  of slag/ash with even higher average radioactivity, is posing considerable environmental problems.

### **PROBLEM: CONCERNS AND THE ASSESSMENT**

#### Increased public pressure

Because of its elevated radioactivity, the landfill has been perceived by the general public in Istria region as a growing environmental threat. The public awareness of the problem is ever more influenced by occasional activist outcries to remove "that high level radioactive waste from the neighborhood". The case of TPP Plomin landfill was even brought to court recently.

The Croatian Electric Utility (HEP), as the TPP Plomin owner, is under an increased pressure to undertake a remediation in order to minimize actual and potential influence of the repository on the local population and the environment. In absence of national regulatory guidance on TENORM management, our Agency (APO-Hazardous Waste Management Agency) was asked to assess the situation and recommend immediate actions aimed at improving containment of the material with elevated radioactivity in the landfill.

### Preliminary safety assessment

APO recommended a simplified safety assessment relying mostly on the data already available. A course of remediation would then be proposed based on transparent reasoning and defensible arguments, starting with affordable interventions, which will result in immediate improvements.

We began with the premise that release of radioactivity to the environment from TPP Plomin landfill during operational phase and post-closure may result in radiation exposure to workers and members of public. Such releases are certainly subject to any criteria applicable to practices in which radioactive material is being handled or waste disposed. In the absence of specific national TENORM regulations, general radiation protection criteria and most relevant international guidance will be followed.

A preliminary safety assessment was performed to estimate occupational exposures, public exposures and environmental impacts. Only operational phase of the landfill was analyzed. Based on the available data, only two normal evolution scenarios were developed for evaluation of the radiological impact and risk posed by the elevated radionuclides concentration in the repository: the first one was "landfill supervisor", describing daily routine of a worker supervising the conveyor operation and unloading of material, and second one was "nearby resident". Other typical scenarios (intruders, disruptive events etc.) were dismissed because of their irrelevance or low probability to occur during the operational phase.

Slag/ash had been used as building material for some of the dwellings in nearby villages. In those places radon-222 levels were controlled by measurements, but this problem was not considered here.

The first scenario assumed that a worker spends 6 hours daily at the landfill: 4 hours in the control shelter and 2 hours on the spot where the waste material is unloaded (being exposed during 75% of the work-time). The 42 hours work-week was assumed. The analysis assumed that all decay products were in equilibrium with parent nuclides. Corresponding irradiation model was based on (a) direct gamma-radiation from the landfill, and (b) airborne releases of radon and fugitive dusts.

The second scenario was considering several populations residing in immediate neighborhood (within 100 m from the landfill boundaries) and at various distances (up to 10 km from the landfill).

Based on the investigation of samples in the vicinity of the landfill, the model assumed that no spread of uranium-238 and radium-226 has yet occurred by leachate migration to groundwater or by surface water runoff to the nearby stream. Therefore only particle transport by wind was modeled, involving (a) impact of direct radiation by precipitated particles, (b) inhalation of particles and (c) consumption of contaminated food. Current analysis did not consider any other transport mechanisms by which radionuclides could move from the landfill and become potential sources of radiation exposures.

This preliminary assessment relies on a rather weak assumption of an average concentration of radioactivity in the landfill. Radioactivity was probably log-normally distributed throughout the volume of repository in the time when only local coals were used. After mixing of coals started, and in particular after the local mines were closed and only coals with low radioactivity were used, the bulk volume is primarily characterized by very low concentrations but with irregularly scattered pockets of increased activity. To better model radionuclides migration in the future assessments, it will be necessary to refine the relationship between waste volume and radionuclide concentrations by direct measurements. There is also a need to evaluate further physical characteristics of the waste. In order to include surface water runoff to nearby streams an leachate migration to groundwater in the next assessment iterations, more reliable information needed for modeling the radiological source terms as well as environmental transport and mobility should be collected. In the future following parameters will have to be re-examined for long term safety assessment: waste permeability, distribution of particle sizes, porosity, hydraulic conductivity, leach rate and waste-to-water distribution coefficient (K<sub>d</sub>).

Simple codes (e.g. RESRAD and PATHRAE) were used for preliminary dose calculations. Conservatively assuming an average specific activity of uranium-238 at 1,500 Bq/kg and of radium-226 at 1,300 Bq/kg for the landfill with uniform distribution of radioactivity (the actual average values are lower), potential dose of 0.6 mSv/y was calculated for the "landfill supervisor" scenario. The biggest portion of the dose is coming from direct gamma exposure from the disposed material, radium-226 being responsible for almost 80% of the dose. Both of the codes generated consistent values within 15% differences. For the "nearby resident" scenario only PATHRAE code was used, and potential dose was calculated to be 60  $\mu$ Sv/y for the

resident spending all of the time within radius of 100 m from the landfill. The calculated values can only be taken as very rough estimate, due to the simplified modeling of the very complex system and the uncertainties in available data.

#### **Regulatory requirements and guidance**

In the absence of clear guidance by national legislation, and preparing for the process of accession to European Union (EU), we adopted harmonized European criteria that can be applied to the TPP Plomin landfill.

Title VII of EU Council Directive 96/29/EUROATOM obliges the member states to investigate the problem of natural sources of radiation, to ensure an extensive monitoring programme and to take adequate measures in case of increased exposure (8). The measures to be taken by member states in interventions where exposure from historic contamination exists, are adopted from the ICRP-60 (9). However, there are no strict guidelines given on measures to be taken by each of the EU member states in intervention situations.

In ICRP-60 doses to members of the critical group are required not to exceed a dose limit of 1 mSv in any year, or not more than 5 mSv in a single year provided that average dose over five consecutive years doses not exceed 1 mSv per year in special circumstances.

On more general issues of radiological protection and safety, the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (BSS) was used as guidance. The BSS adopt the effective dose definition by ICRP-60, with primary dose criteria for the unconditional exemption of material from regulatory control of 10  $\mu$ Sv/y above background to an average individual in a critical group (10).

Our preliminary safety assessment results are clearly indicating the need that the repository should be put under regulatory control, although the dose for the worker is not exceeding the 1 mSv per year limit required for intervention. Once under regulatory control, TPP Plomin slag/ash landfill should be operating in a way which ensures that protection of the public and the environment is optimized, doses being kept as low as reasonably achievable (ALARA) and both social and economic factors taken into account.

A protecting system based on engineering barriers or other measures should be used to attain a level of radiological protection that meets the dose or risk constraint which can be derived from the ICRP recommendations or IAEA Safety Series (the suggested values are from 0.1-0.3 mSv/y). In the future these constraints will be determined by Croatian national regulatory body. Combination of engineering and institutional controls should give reasonable long-term assurance that containment of the material and increased radioactivity in landfill will remain effective for the TPP operating period as well as eventually in the post-closure period.

#### THE IMMEDIATE RESPONSE

Recently, a remediation project for the TPP slag/ash landfill has been initiated, prior to the expected national regulatory developments aimed at TENORM management. The project goal is

to immediately improve conditions at the site. Appropriate barriers should be engineered in order to lower the doses to the workers on the landfill, and in order to improve containment of the waste so as to reduce transport into the environment, in particular towards the Plomin bay. Guidance for the basic design was sought both in the European (11) and in the national regulations on inert waste landfills. The regulations specify that the protection of soil, groundwater and surface water is to be achieved by a combination of geological barrier and bottom liner during the operational phase, and by a combination of geological barrier and top liner during the post closure period.

This remediation project is conceived as a prompt but partial solution. After its completion, comprehensive measurements will be carried out on the improved landfill in order to collect data on all relevant parameters, in particular about the actual heterogeneous distribution of the radionuclides concentrations in the waste. A new iteration of safety assessment, based on these data and describing future performance of the improved landfill in appropriate detail and with greater resolving power, should provide more realistic estimates of the maximum expected doses to the workers, of the potential exposures in the neighboring population, and of the potential impact on the environment. The next phase of remediation will depend on the assessment results, which should indicate if any intervention in the waste layers or some new engineered barriers will be needed to improve protection during operational period and to ensure long term safety in the post closure period.

Taking into account on-site dose measurements and radioactivity of the samples from the bay and from the landfill surroundings, and the results of the preliminary safety assessment, as well as the regulatory requirements for such landfills, remediation activities were proposed and have already started, with the primary aim to reduce the potential for transport of radionuclides into the environment by:

- (a) preventing dust transport,
- (b) minimization of leaching, and
- (c) preventing materials washout.

A remediation project complying with the detailed requirements of the regulations mentioned above should:

- (a) geomechanically stabilize the landfill, and in particular construct 1:3 slopes around the perimeter;
- (b) level the surface and build at least 1 m thick top soil cover growing grass (extending over the slopes), except at the portion where new waste is brought in;
- (c) ensure that torrents cannot approach the landfill;
- (d) prevent dust production and spreading by wind;
- (e) engineer drainage of the landfill surface;
- (f) ensure the drainwater collection and sedimentation before it is released to the sea;
- (g) the landfill base and sides should consist of a mineral layer which, combined with the geological barrier, satisfies permeability limit of at most 10<sup>-7</sup> m/s and is at least 0,5 m thick.

To achieve that, in the year 2001 we plan to:

- (a) build a fence sufficient to prevent free access to the site;
- (b) along the sides of the valley build a perimeter canal to channel torrents, especially the Bizac torrent;
- (c) build a canal network on the landfill cover with a sedimentation collector for the precipitation water from the cover;
- (d) arrange for enclosed transport of the materials to be deposited in the landfill. The remediation will be supplemented by the systematic radiological monitoring of conditions on the site, and supported by a new strategy of materials depositing, designed to increase geomechanical stability of the landfill and level its surface with a layer of new slag/ash of low radioactivity produced by the coal now in use.

At present, the engineering of bottom and side liners remains an open issue. Lifting and moving 900.000  $\text{m}^3$  of waste in order to build 0,5 m thick  $10^{-7}$  m/s permeability mineral layer is obviously a complex operation, which should be justified by a careful evaluation in spite of the formal requirements. Prospective advantage of the improved radioactivity containment, and the opportunity to collect and monitor the leacheate, are opposed by the foreseeable generation and uncontrollable dispersal into the environment of large quantities of radioactive dust during a prolonged period of excavation, transportation and redisposal of the entire waste. A detailed assessment of possible radiological consequences, as well as a cost-benefit analysis, should be carried out before the final decision is made. For the long lived radionuclides in the waste, any bottom liner is but a short term safety feature. It perhaps can be compensated for by specific cover engineering (including e.g. an artificial sealing liner) and by appropriate disposal procedures designed to reduce rain infiltration during the operational phase of the landfill.

### CONCLUSION

The results of a recent preliminary safety assessment indicate that the TPP Plomin slag/ash landfill contains TENORM, which cannot be automatically exempted from regulatory control according to the BSS recommendations. On-site measurements have not revealed any appreciable migration of radionuclides into the environment, but a relatively simple remediation has been started in order to reduce potential for atmospheric transport, washout and leaching.

The choice to remediate promptly, as well as the decision on appropriate level of the present remediation, was motivated by the prospective benefits in terms of potential doses reduction for those who work on the landfill or on the nearby land, and by the cost of remediation. In addition to radiological consideration, the decision to remediate was also influenced by the public concern and anxiety.

Improving the physical protection of the landfill, the partial cover construction and other measures against rain and surface water infiltration will alleviate the present radiological problem, both for the workers engaged in waste disposal and for the neighboring population. For the next step, however, better regulatory guidance will be needed, particularly because any further intervention should primarily be aimed at the safe use of the location in the post closure period and the long term safety. Since there are no national regulations on TENORM, they

should be introduced in the meantime. Regulatory body should allocate annual dose constraints for the landfill in the operational and the post closure periods, which would ensure that overall dose limit is not exceeded, taking into account releases and exposures from all other relevant sources and practices.

Despite public concern, the immediate radiological problem is not really a big one. But because of the very slowly decaying radionuclides, its long term character must be acknowledged. Therefore, any further remediation activities should be based upon a thorough safety assessment, particularly focused on the landfill post closure performance. As this preliminary iteration has indicated, present data on the materials disposed are by far insufficient. Some of the uncertainties can be reduced though not eliminated by better landfill characterization, by using refined models or by obtaining more site specific data. Only after that a qualified decision can be made about possible benefits of additional engineered structures in the landfill.

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