# NORM AND TENORM IN AUSTRIA

*F. Schönhofer, Federal Ministry of Agriculture, Forestry, Environment and Water Management, Radiation Protection Department, Vienna, Austria*

## 1 ABSTRACT

According to Title VII of the European Union Radiation Protection Directive 96/29 EURATOM the member states have to perform investigations about occurrence and possible impact of naturally occurring radioactive material. In the case of for instance radon in indoor air and NORM in drinking water studies exist. To identify possible sources in industry and and to estimate, whether a possible risk might be associated a survey was done in Austria. The strategy and the outcome are described in this paper.

## 2 INTRODUCTION

Among naturally occurring radioactive material the radionuclide K-40 and some members of the natural decay chains (Rn-222, natural uranium, natural thorium, Ra-226, Ra-228, Pb-210 and Po-210) are of interest as potential causes for elevated doses to workers and members of the public, which cannot be disregarded from the radiation protection point of view. There are natural, unprocessed substances known, which contain elevated levels of these radionuclides (“NORM”). On the other side, processing of certain substances (e.g. ores) might result in technological enhancement of NORM (“TENORM”).
Title VII of the Directive 96/29 EURATOM (1) states, that “each Member State shall ensure the identification, by means of surveys or by any other appropriate means, of work activities which may be of concern”. Dealing with NORM and TENORM the exemption values for practices set out in both column 2 and 3 of Table A to Annex 1 for reporting have to be considered. The respective values for the most important radionuclides in NORM are summarized in Table 1. Austria has not yet set clearance levels according to Article 5 (2).

Table 1: Exemption values for practices from (1) for NORM radionuclides

| Nuclide | Activity (kBq) | Activityconcentration (kBq/kg) |
|---|---|---|
| K-40 | 1000 | 100 |
| Pb-210+ | 10 | 10 |
| Po-210 | 10 | 10 |
| Ra-224+ | 100 | 10 |
| Ra-226+ | 10 | 10 |
| Ra-228+ | 100 | 10 |
| Th-232sec | 1 | 1 |
| U-238+ | 10 | 10 |
| U-238sec | 1 | 1 |

The European Commission has published several documents, recommendations and guides on NORM, TENORM and the implementation of Title VII into national law (2, 3, 4 and 5). Though these references contain much information about potential sources, these sources are of limited use for Austria, since mining and industrial activities have to be considered from case to case. Furthermore the compilation is not exhaustive.

The strategy of the survey in Austria was first to compile the information, which was already available from previous research, then to check what kind of industry exists in Austria, which is known to may be affected by NORM or TENORM and finally to identify practices with potential risk for elevated radiation doses, which are usually not considered and are hardly or not at all mentioned in the literature.

As sources and potential sources for elevated doses the following practices and industries in Austria were identified:

Radon exhalation from soil and accumulation in houses and working places,
radon exhalation from water and accumulation in water works,
radon in mining and excavation,
mining and milling of ores
phosphate and fertilizer industry
coal mining and burning
oil and natural gas industry
rare earths industry
zirconium industry
radium industry
thorium industry
purification of water.

The results of the survey will now be discussed for the potential sources.

## 3 RADON IN AIR

The most important source of radon in indoor air is the soil beneath the house. Further sources may be degassing from radon bearing water and exhalation from building material, but their contribution to the overall radon concentrations in indoor air is usually insignificant.

Since 1992 a systematic survey of radon concentrations in indoor air of private houses was financed by the Austrian Radiation Protection Authorities and carried out by a group of Austrian scientists and laboratories under the coordination of the Institute for Isotope Research and Nuclear Physics. A summary of the results can be found in (6). One of the outcomes of the study is a “radon potential map” (Fig. 1), from which easily areas with elevated risk for high radon concentrations in indoor air can be identified.

During the year 2000 a comprehensive survey on radon in kindergartens was carried out in the province of Upper Austria (7), a survey of school is to follow.

Also in Upper Austria a project on radon mitigation methods was undertaken (8).

The information obtained from these surveys, from the Austrian Radiation Atlas (9) and the information on radon in groundwater (see below) will simplify the search for work places with enhanced radon concentrations.

## 4 RADON IN WATER

Since the beginning of the 20$^{th}$ century radon concentrations in many kind of Austrian waters were measured by the Institute of Radium Research. In the 90's two systematic studies in the provinces of Lower Austria and Upper Austria were carried out, which were directed toward the concentrations of radon and radium in drinking water. The results can be found in (10) and (11). The available data were combined to a radon potential map for water (12).

The contribution of radon from water to the radon concentration in indoor air is usually small, but still can give rise to an additional, not neglectable dose. However, in the case of water works with its large pools of water, radon concentrations in air might rise to extremely high values. Therefore measurements were undertaken in the drinking water supply system of a very big Austrian town (13). Depending on the origin of the water values from 20 to 800 Bq/m3 were measured in the air above the stored water. In no case the water originated from a risk area, most came from karstic areas and had very small concentrations in the water. It can be assumed, that in risk areas the values would be dramatically higher. This is no reason for health concern, because the workers in the plants spend very little time inside the water halls, usually only for controls and for repairs.

## 5 RADON IN MINING AND EXCAVATION

Since 1970 measurements in underground mines, tunnels and in spas have been carried out (14). In 50% of underground mines 0.3 WL were exceeded, which corresponds to about 15 mSv per year. Also in tunnels and in hotels in spas values exceeding 0.3 WL were observed, though the mean values were generally below 0.1 WL. These findings resulted in mitigation measures like improvement of air exchange and removal of radon from water before entrance into the hotel.

## 6 MINING AND MILLING

The highest potential for elevated doses from NORM is in uranium mining. Since in Austria no economically exploitable uranium ore deposits are known, there is no uranium mining.

Many ores contain uranium and thorium as well as their daughter products in broadly varying concentrations. During milling they may be liberated or concentrated in byproducts. Pb-210 and Po-210 are volatile and maybe condensed in tubes, concentrated in fly-ash or washed out during off-gas treatment. Fly-ash may be deposited and radionuclides as well as heavy metals may be washed out and reach the ground water table.

There is a trend to use byproducts commercially, in order to save costs for disposal. Slag and fly-ash is used in construction material. Slag, ashes and other residues from the milling industry often contain not only TENORM, but other metals and they are used for extraction of these other metals, which might yield other residues with even higher concentrated radionuclides (Table 1).

Table 2: Import of ores, concentrates, slags, ashes and residues into Austria, 1999 (15).

| | |
|---|---|
| Aluminium ores and concentrates | 26 336 t |
| Lead ores and concentrages | 867 t |
| Titanium ores and concentrates | 104 t |
| Zirconium ores and concentrates | 1 017 t |
| Vanadium ores and concentrates | 873 t |
| Titanium oxide | 2 770 t |
| Vanadium oxide | 2 752 t |
| | |
| Granulates slag from iron and steel manufacturing | 34 890 t |
| Ashes and residues, containing mainly lead | 3 910 t |
| Ashes and residues, containing mainly copper | 896 t |
| Ashes and residues, containing mainly vanadium | 879 t |

Only few minerals are mined nowadays in Austria, which theoretically may be of concern for workers: Coal is mined in two open strip mining operations – therefore very little potential exists for elevated doses, iron ore is mined in one underground mine and one open strip operation, tungsten is mined in two underground mines. Out of geological considerations it has to be expected, that no significant thorium and uranium concentrations are associated with the tungsten ore mined.

Altogether there are 23 underground mines in operation in Austria. These have to be considered as potentially exhibiting elevated concentrations of radon. Some work in this respect has been done (see Chapter 4) already. Further investigations are to be carried out in the planned survey.

# 7 FERTILIZER INDUSTRY

Raw phosphate rock contains uranium, thorium and radium in concentrations, which depend on the origin of the rock. The most frequent method to produce fertilizer is treatment of phosphates with sulphuric acid, which yields phosphoric acid for fertilizer production and gypsum as a byproduct. Uranium and thorium are concentrated in the phosphoric acid, most of the radium is precipitated together with gypsum, but still is present in the phosphoric acid also. Only a small part of gypsum can be used commercially, most of it has to be deposited on piles. These piles show elevated gamma-doserates and they emanate radon. For the use of phosphogypsum in agriculture the radium content has to be considered.

Potassium salts are also a constituent of fertilizers, therefore they contain both Ra-226 and K-40.

There are many pathways for exposure from the fertilizer industry. Within the phosphoric acid plants workers may be exposed to significant doses due to external radiation from radium rich scales, inhalation doses are caused mainly by Po-210, but also Rn-222. From the phosphogypsum piles uranium, Po-210 and Pb-210 and little Ra-226 may be washed out and reach the groundwater or aquatic ecosystems. Solid waste can be filtermaterial, scales and dust, enriched in radionuclides. During storage and packing workers may be exposed to external radiation (both from radium and K-40), inhalation of radon and inhalation of dusts cause internal radiation of the lung.

Members of the public may be effected by phosphogypsum used for house construction purposes both by gamma-radiation and exhaled radon.

In Austria two companies existed, which produced fertilizers and used the sulphuric acid method. One of them used to manufacture building construction material from the by-product phosphogypsum. A few years ago this company has ceased manufacturing fertilizers and uses gypsum which is a by-product from the manufacture of citric acid. Until now, no information is available on the origin of raw phosphate rock used by both companies or on the activity of the mentioned by-product gypsum. There is reason to suspect, that significant doses are associated with the processing of phosphate rock and handling of fertilizer, because analysis of grab samples of Austrian fertilizers showed concentrations of up to 1 500 Bq/kg Ra-226 and 8 500 Bq/kg K-40 (16). Regarding the situation of gypsum used in house construction it is known, that about 20 years ago plates of gypsum showed sometimes elevated levels of gamma radiation. On the other hand there is information, that one large manufacturer of plaster boards uses exclusively natural gypsum, which is practically free of Ra-226.

These considerations will govern the strategy of the planned survey with respect to fertilizers, since it cannot be excluded, that the fertilizer industry might be a source of elevated exposure both for workers and members of the public.

## 8 COAL MINING AND BURNING

Coal may contain comparably low levels of uranium, thorium and radium. Water in mines may show high levels of Ra-226, Ra-228 and Ra-224 as has been reported from Poland (17). After burning, most radionuclides remain in both coal ash and fly ash. Volatile radionuclides can be condensed in pipes or emitted over the stack. Ash may be deposited or used in manufacture of concrete and in road construction.

Possible paths of exposure for members of the public are wash-out from ash deposits followed by contamination of ground water, as well as external radiation and radon exhalation from building material with an ash or slag component.

In Austria no exposure pathway can be excluded, but with the exception of some work on the emission of radionuclides from coal-fired power plants no systematic investigations are known. They will be carried out in the framework of the planned survey.

# 9 OIL AND NATURAL GAS INDUSTRY

Water associated with oil contains little U-238 and Th-232, but usually clearly elevated concentrations of Ra-226, Ra-228 and their daughter products. During oil pumping and separation of natural gas and water changes in pressure and temperature occur, which causes radium to precipitate – with barium preferentially as sulphate, with calcium as carbonate. In pipes and other parts scales are formed, which contain high concentrations of radium and daughter products. Also sludges usually contain elevated concentrations.

Natural gas is always accompanied by radon, the daughter products of which plate out inside the gas pipes and the pumps. During production of liquid petrol gas radon is accumulated in it and the daughter products plate out on the inner wall of tanks and in pumps.

Paths for radiation exposure are external radiation from scales and additionally inhalation of liberated particles during cleaning of equipment. Using high pressure water cleaning the waste water has enhanced radium concentrations, the soil may be contaminated. The scale is in many countries regarded as radioactive waste, which has to be handled and disposed of accordingly.

In Austria no investigations about the situation in the oil and natural gas industry are known, but it is rather likely that this problem is present. Workers might be affected, but members of the public sure are not. It has been found, that natural gas distributed to households in Vienna contains some radon, which originates from the deep geological storage places, where imported natural gas is stored (18). However, the concentrations are far too low to have a significant impact in the household.

# 10 RARE EARTH INDUSTRY

Rare earths are abundant in some minerals like monazite or bastnaesite, where they are accompanied by large amounts of thorium, but also by uranium and radium. Exposure to gamma radiation and dust will be of concern during mining and handling of the ore and especially during isolation of the rare earths.

Austria has a rare earth industry, founded by the famous chemist Auer von Welsbach. Nowadays no raw material is imported any more, but concentrates of rare earths, which contain only traces of thorium.

Due to this fact significant exposure of workers is most unlikely and exposure of members of the public can be excluded due to the extremely small amounts of rare earths used in high tech and every day applications. The company has a policy of demanding strict limits for thorium in the raw products and controls all batches delivered rigorously.

## 11 ZIRCONIUM INDUSTRY

Zirconium ores contain uranium, thorium, Ra-226 and Ra-228. Zirconium silicate and zirconium dioxide respectively are used for the fabrication of refractive material, glazes, glasses and ceramic material, the metal itself is used in special alloys.

Radionuclides are enriched in sludges and by-products. Exposure is possible from storage of raw material and products, dusts may be inhaled.

It is more than likely, that zirconium sands are used in Austria. At least one company manufactures alloys, which contain zirconium. The planned survey will have to carefully consider the zirconium industry.

## 12 RADIUM INDUSTRY

Radium-226 is present in uranium ores. 10 t of uranium ore with 50% uranium oxide yields about 1 g radium-226. Since 1898 the world-wide production was about 4.5 kg radium. The use of radium in luminous paint and in medicine is only of historical interest, though residues at former production and usage places might still cause exposure.

On todays Austrian territory there was only one factory on the outskirt of Vienna which produced radium, using wastes from uranium production in Joachimsthal. Another one produced items containing radium like radium toothpaste, radium bathing soap etc.

According to J. Braunbeck (19), there are no traces left from the latter factory and at the two places in Vienna, where radium was sold, only marginally elevated gamma radiation can be detected.

Exposures might still be possible from medical use of radium. There were a number of rooms in hospitals for radium therapy, which were highly contaminated by radium, but also be Pb-210, which plated out from radon decay. As far as it is known, all such rooms have been decontaminated already.

Frequently items are found, containing luminous paint on the basis of Ra-226, like in old airplanes and locomotives. Most of them and other consumer products like watches are not used any more and therefore pose practically no risk for significant exposure.

It was not possible within the framework of this preliminary study to find out, whether a watch industry existed in Austria, using radium-paint. This seems to be the only question open for the planned survey with regard to the radium industry.

# 13 THORIUM INDUSTRY

Historically seen the main use of thorium was in the form of thorium nitrate for the production of gas lantern mantles. Today there exist non radioactive alternatives, which use mostly yttrium nitrate. Thorium metal is used as a hardener for magnesium alloys, which are used for instance for air craft motors. A tungsten alloy with 2% of thorium is used for welding electrodes. Thorium oxide is a constituent of optical glasses and refractive material. Finally thorium is present in many minerals and is due to its radioactivity an unwanted impurity in many industrial products.

According to (15) 1200 kg of “thorium compounds, in U235 depleted uranium” was imported into Austria in 1999. Of course no statistic is available, how much thorium was imported due to impurities of other minerals.

There has been a production of thoriated gas lantern mantles in Vienna, but the company has replaced thorium by yttrium a few years ago. Thoriated tungsten welding electrodes are produced in Austria, but the company advertises another type of electrodes as an alternative as well, stating that they do not have the “drawback of being radioactive”.

# 14 WASTE FROM PURIFICATION OF WATER

Water, especially ground water contains natural radionuclides, the concentrations being dependent on the origin of the water. Maximum permissible concentrations, respectively the maximum permissible dose due to incorporation of radionuclides from drinking water are given in the Drinking Water Directive 98/83 EG (20). Many methods are known for removal of NORM from water. A comprehensive research project was funded by the European Union (21). The waste (ion exchange resins, granulated activated carbon, waste streams from reverse osmosis, sludges etc.) contain most of the radionuclides. But methods for removal of other unwanted non-radioactive constituents will remove in many cases also radionuclides, which may be present in concentrations far below the maximum ones according to Directive 98/83. They will be concentrated to high activity concentrations and the total activity will be very high as well, because of the high amount of water purified.

It has been shown also by field measurements, that for instance the removal of iron and manganese by oxidation and precipitation of the oxide hydrates automatically removes most of the radium. Waste from purification of waters high in iron and manganese might even in cases with relatively low radium concentration in water become radioactive waste (22).

The above given considerations concerning radium apply to other radionuclides like Pb-210 and uranium as well, since they are removed easily by many purification methods. An overview on removal efficiencies for various nuclides is given in Table 3.

Table 3: Removal efficiencies for radium, lead and uranium

| treatment method | Ra | Pb | U |
|---|---|---|---|
| GAC Filtration | 0 – 90 | 30 – 99 | 0 – 99 |
| Cation Exchange | 95 – 98 | 20 – 90 | - |
| Anion Exchange | 35 – 60 | 20 – 70 | > 95 |
| Mixed Bed Exchanger | 95 – 98 | 20 – 90 | > 95 |
| Hydroxylapatit Filtration | 35 – 95 | 75 – 95 | 58 - 82 |
| Reverse Osmosis and Nano Filtration | > 95 | > 95 | > 95 |

Unfortunately these considerations have not received much attention yet, though the disposal of waste from water purification might in many cases pose a difficult problem. In Austria there is a potential for elevated radium concentrations in water (10) and the risk areas are known (Figure 1, Figure 2). The planned research project on NORM and TENORM in Austria will have to deal with this subject.

## 15 CONCLUSION

This preliminary survey has identified industries and sources of NORM and TENORM which might pose a risk of significant exposure to workers or members of the public. It was found out as well, that some industries, substances and products which are of concern in other countries are not present in Austria or play only a negligible role and can be disregarded. This preliminary study, which used available information from import statistics, industrial production methods to available measurements is therefore the basis for comprehensive research and the necessary measurements to fulfill the requirements of the European Union Directive 96/29 EURATOM, Title VII.

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