Four decades of radioecological research on the Danube river: a reliable basis for protecting the freshwater resources of central Europe of tomorrow

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INTRODUCTION

Since the beginning of the atmospheric nuclear weapon test period in the end of the fifties the radioactive contamination of the environment was monitored and evaluated by scientists of national health authorities in regard to minimise the health risks of the population. The Danube with a total length of 2857 km and a drainage area of about 817,000 km² in central Europe (Fig. 1) became also an important ecological target for monitoring and evaluation of man-made radioactivity.

As the second biggest river in Europe after the Volga the Danube is used as drinking water source, for irrigation purpose, for fishery, for sport and recreation, as water supply for industry, and for transportation. So the Danube acts as live resource for nearly 70 million people in Europe.

Figure 1. The course of the Danube in Europe and Austria

In the upper part of the Danube, from its springs in the Schwarzwald in Germany to the Hungarian lowland, the hydrological characteristic of the river is dominated from the Alpine morphology and climate. In this section the river has got a very widespread variation of flow rates and suspended matter loads. The long-term annual mean flow rate in Vienna is about 1900 m³/s. Downstream the river is developing in a more calm and well-balanced large river with a mean flow rate of about 6600 m³/s (1).

The biggest tributaries of the Danube with major increase of discharge are the rivers Inn, Drava, Tisa, and Sava (Fig. 2). The mean slope of the Danube in the upper part - km 2880 to km 1800 is: 0.2 m/km, in the mean part - km 1800 to km 950: 0.08 m/km, and in the lower part - km 950 to km 0: 0.06 m/km.
Figure 2. Hydrographic profile of the Danube from its spring to its delta at the Black Sea [1]

An important role in the transportation process of pollutants like heavy metals or radionuclides play solid particles and sediments. Due to the weathering process in the Alpine region the main part of the suspended material in the upper part of the Danube are mainly (95% to 99%) mineralogical components and less organic particles. The main characteristics of the water and the sediment of the Danube’s upper part is given in Tab. 1.

Figure 3. Sum frequencies of measured suspended matter concentrations of the upper part of the Danube (1989-1992, raw data: Wasserstrassendirektion, Vienna)
The relatively high clay fraction of the sediments leads to a high binding capacity of solid particles in the Danube's water for cationic pollutants like radio-caesium. In connection with the lively hydrographic characteristic of the Danube's upper part the annual main turnover of ecological radioactivity happens during flood events some days the year. In Fig. 3 the wide variation of suspended matter concentration in the Danube water (Austrian part) is shown. The measured extreme values of suspended matter concentration are < 2 mg/l in low water situation to 13000 mg/l during extreme flood situations.

In the mean and lower part of the Danube the hydrological regime changes and the share of organic components of suspended matter increases. Therefore metal-complex binding processes come to the fore. Whereas in the inorganic mineralogical clay dominated solid phase the remobilization of particle bound radionuclides is very low the exchange between the water, suspended mater and biological compartments are generally higher in organic dominated fluvial environments. So in the mean and lower part of the Danube the biological availability of some particle bound radionuclides generally increases.

### Table 1. Water end sediment characteristics (mean values) of the Danube (upper part)

<table>
<thead>
<tr>
<th>Water - chemical characteristics</th>
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<tbody>
<tr>
<td>pH</td>
<td>7.5</td>
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<tr>
<td>electr. conductivity</td>
<td>250 µS/cm</td>
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<tr>
<td>Ca$^{2+}$ (CaCO$_3$, Ca(HCO$_3$)$_2$)</td>
<td>50 mg/l</td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>0.4 mg/l</td>
</tr>
<tr>
<td>PO$_4^{3-}$</td>
<td>0.2 mg/l</td>
</tr>
<tr>
<td>NO$_3^-$</td>
<td>5 mg/l</td>
</tr>
<tr>
<td>AOX</td>
<td>20 µg/l</td>
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</table>

<table>
<thead>
<tr>
<th>Sediments - mineralogical composition</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>clay and mica minerals</td>
<td>44 %</td>
</tr>
<tr>
<td>quartz and feldspar</td>
<td>30 %</td>
</tr>
<tr>
<td>carbonates</td>
<td>22 %</td>
</tr>
<tr>
<td>organic matter</td>
<td>4%</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Sediments - grain size distribution</th>
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<tbody>
<tr>
<td>Clay fraction (&lt; 2 µm)</td>
<td>15 %</td>
</tr>
<tr>
<td>silt fraction (2 - 63 µm)</td>
<td>70 %</td>
</tr>
<tr>
<td>fine sand fraction (63 - 250 µm)</td>
<td>15 %</td>
</tr>
</tbody>
</table>

The most important sources of artificial radionuclides in the Danube over the last forty years are the atmospheric nuclear weapon tests and the environmental contamination after the Chernobyl accident. In a lower extend the nuclear power plants in the Danube's catchment area in Germany, Czech, Slovakia, Hungary, Bulgaria, and Rumania contribute to the man-made radioactivity in the Danube.

In this paper a comprehensive review on some important results of radioecological research work done at the Danube in four decades is given as a basis for future research work and ecosystem management decisions.

### MATERIALS AND METHODES

According to the development of the environmental radioactivity and the public interest on the topic three main investigation periods of Danube radioecology research could be summarised in the last four decades: The early phase from the beginning of the sixties to the middle of the seventies, the middle phase after the first international oil crisis and before the Chernobyl accident, and the late period after the Chernobyl accident till now. The investigation targets in this three main periods have been developed from a monitoring level in the early phase to a more public dose and health risk modelling oriented period in the middle phase. In the late phase the total ecological connections of environmental radioactivity have been given special emphasis.
In parallel to the scientific and public interest on investigations of environmental radioactivity the technology stage of radio-analytical methods increased rapidly. In the early period total-beta and total-alpha measurements of Danube water, biotic samples, and fish was carried out by gas filled proportional counters. NaI(Tl)-scintillation detectors had been used for gamma spectrometric analysis of water and biological material. After the early period the technological development of liquid scintillation counters for beta and alpha radiation and semiconductor detectors for alpha spectrometry and gamma spectrometry led to nuclide specific activity analysis.

The need of a certain scientific basis for decisions on measures of the public health authorities after the widespread radioactive contamination of the environment as a result of the Chernobyl accident called for new and practicable models, parameters and managing tools. This necessity has promoted an amount of research work to understand the connections of environmental compartments on the behaviour of radionuclides in the Danube ecosystem.

RESULTS

The first radioactivity monitoring and investigation program of the Danube was co-ordinated by the International Association for Danube Research IAD. The scientists of the riparian countries along the Danube from Germany to the Soviet Union measured and evaluated between 1958 and 1965 water, sediment and river organism in order to monitor the man-made radioactivity in the river and to evaluate health hazards and risks by using the water and fish of the Danube. As mentioned before in this early period of radioecological investigations the total-beta-activity and the residue-beta-activity (beta-activity without K-40-activity) was examined mainly by means of proportional counters.

The maximum values of artificial radioactivity was found in the samples taken in the year 1963. These peak values were caused by the large amount of atmospheric nuclear weapons tests of the superpowers. In 1963 the maximum residue-beta-activity of Danube water between Germany and Rumania was about 600 mBq/l (2). The maximum values of total-beta-activities in sediment-ash were about 3700 Bq/kg and in green algae-ash around 32000 Bq/kg (2).

Ten years after the maximum atmospheric nuclear weapons test series a Rumanian researcher (3) analysed the long-lived radionuclides Cs-137 and Sr-90 in the water of the Rumanian part of the Danube. He found Cs-137 activity concentrations between 7 mBq/l to 17 mBq/l and Sr-90 activity concentrations from 3.7 mBq/l to 400 mBq/l in Danube water samples.

A Bulgarian researcher team reported results of radioactivity measurements of water and fish samples of the Bulgarian part of the Danube between 1964 and 1974 (4). They found Sr-90 values in water of about 30 to 100 mBq/l and Cs-137 in water of about 2.2 mBq/l. In fish (barbus barbus) they measured Sr-90 activities of about 250 mBq/kg and Cs-137 activities of about 230 mBq/l.

After this first period of radioecological work at the Danube a more comprehensive and public dose focused approach was taken by the scientists. In this second period from the middle of the seventies after the first oil crisis to the middle of the eighties (before the Chernobyl accident) new radioecological transfer models and the collective dose concept had been applied. In this period an international 4-years (1976-1979) co-ordinated research programme of the IAEA on the problems of the radioecology of the Danube river was carried out (5, 6). The generally decreasing level of atmospheric and hydrospheric artificial radioactivity in this period after the implementation of the nuclear test ban caused only nearly negligible contributions to the average annual dose of the population (5, 6, 7). The implementation of transport models and compartment models on the Danube for activity prediction and dose assessment promoted fruitful work to understand the radioecological processes and the relevant parameters and connections (8, 9).

The routine release of radioactivity from nuclear power plants in the Danube's catchment area did not contribute to a significant public dose in this period.

The third period of investigation on the radioecology of the Danube has been strongly influenced by the contamination of the environment after the accident of Chernobyl. In the course of managing the environmental consequences of the nuclear accident all of the national radiation protection authorities in Europe recreate former or create new radioecological monitoring and evaluation programs. So the Danube as one of an important lifeline of the central European countries became again an interesting object for radioecological research, protection and restoration. In this period a permanent international grab sampling and radiometric analysis network along the Danube within the scope of the International Association of Danube Research IAD and the national authorities of the riparian states has been installed.

In the first weeks after the reactor accident the radioactive contamination by Cs-137 and Sr-90 of water, suspended matter, and sediment of the Danube increased by factors between 10 and 1000 dependent on the radionuclide and the compartment (10). Some hundred reports and scientific publications related with
radioecological and radiation protection topics in connection with the Danube has been published since the Chernobyl accident so far. Therefore only some results of long-term monitoring programs and some fundamental results could be presented in this paper.

In Fig. 4 the long-term activity concentration of Tritium in Danube water is shown. The H-3 activity concentration decreases from about 200 Bq/l in the early sixties to about 1.5 Bq/l in the end of the nineties. According to these results there was no influence of the Chernobyl accident on the H-3 regime of the European hydrosphere.

Figure 4. Long-term course of Tritium in Danube Water (km 1931, Vienna) and in precipitation (11)

Figure 5. Long-term course of Cs-137 activity concentration of sediment samples of the Austrian part of the Danube
The development of the Cs-137 activity in Danube sediment from 1986 to 1998 is shown in Fig. 5. In autumn 1986 the Cs-137 activity concentrations of Danube sediment were between 1000 and 2000 Bq/kg. In the first year after the Chernobyl accident the ecological half-life in the sediment compartment of the Danube was about 5 month. Since 1987 an ecological half-life for Cs-137 in Danube sediment of about 4 years could be observed. In the end of the nineties the C-137 activity concentrations were between 50 and 100 Bq/kg (Fig. 5).

For Cs-137 there is a strong dependency of mass specific activity concentration to the grain size distribution of the sediment (Fig. 6). The binding affinity of Cs-137 on clay particles (< 2 µm) is about a factor of 10 higher then for sandy particles (> 100 µm). This behaviour effects strongly the Cs-137 activity concentration of sediment samples. The Cs-137 activity concentration depends on the grain size distribution of the sediment. This fact have to be taken into account by using analysed values for further radioecological models and evaluation (12, 14, 15, 16).

Figure 6. Dependency of the activity concentration on the grain size of Danube sediment

Figure 7. Distribution of activity concentration of water samples, Danube Austria (distribution of analysed values in box-plot with 5%-10%-25%-50%-75%-90%-95%-quantile)
Figure 8. Seasonal variation of Cs-137 and Pb-210 excess activity concentration of Danube sediment
(distribution of analysed values in box-plot with 5%-10%-25%-50%-75%-90%-95%-quantile)

In Fig. 7 the measured activity concentration frequencies of natural and artificial radionuclides of
Danube water samples are summarised. The analysed values, represented as box-plots, indicates the activity
concentrations of the solved phase and of the particle bound (solid) phase of each radionuclide in the period 1993
to 1998. In average the particle bound transport of Cs-137 is about three times higher than the solved transport.
During flood events in the upper, alpine dominated part of the Danube the particle bound portion of the Cs-137
activity could be more than 1000 times higher than the solved part. Therefore in average in less than 30 days of a
year more than 90 % of the Cs-137 activity load is transported bound on particles (suspended matter) in the
Danube.

The general seasonal trends of activity concentration for Cs-137 and Pb-210 excess in Danube sediment
samples are shown in Fig. 8. During summer the activity concentrations are generally lower and during winter the
activity concentrations are generally higher than the average values of sediments. This behaviour is based on
stronger weathering processes in the alpine catchment area during summer combined with lower activity solid
particles and enrichment processes due to erosion of more fine grained, higher activity particles during winter.

Figure 9. Mean Cs-137 activity concentration in Danube fish, Hungary (13)
As an example for the radioactive contamination of biotic compartments the time course of mean Cs-137 activity concentration of various fish samples, taken in Hungary are given in Fig. 9. In the end of the nineties the Cs-137 activity concentration have decreases in average below 1 Bq/kg.

CONCLUSIONS

The long-term course of the man-made radioactive burden of the Danube in the last four decades was highly affected by two incidents in the northern hemisphere: the atmospheric nuclear weapon tests of the superpowers in the late fifties and the early sixties and the environmental contamination following the nuclear power plant accident of Chernobyl in 1986.

In regard to the activity load of Cs-137 and some other artificial and natural radionuclides transported in the water of the Danube the suspended particles combined with flood situations are of great importance. In the upper, Alpine influenced part of the Danube more than 90 % of the annual Cs-137 activity, washed in the waterways of the Danube's catchment area by erosion processes during precipitation events, is transported in less than 30 days of the year.

Based on radioactivity measurements of thousands of samples of all the compartments (water, suspended matter, sediment, biotic materials, organisms) of the Danube done and reported in the last four decades no general evidence for significant health risk due to man-made radioactivity could be found. But during some months after the maximum nuclear weapons test period in 1963 and during the first weeks after the Chernobyl accident some compartments and organisms of the Danube were contaminated with elevated levels of various artificial radionuclides. In these periods significant dose contributions for small population groups along the Danube via fish consumption or agricultural use of Danube water for irrigation could not be excluded absolutely.

The improved and well-evaluated knowledge of the radioecological behaviour of man-made radionuclides in the catchment area and the compartments of the Danube during the last four decades could be a reliable scientific basis for protecting this European life-line in the future. But only responsible behave and careful management and circumspect use of the environment resources by men could maintain and protect the special and unique hydrosphere and special biosphere of the Danube permanently.

REFERENCES

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