

SHORT COMMUNICATION

CHERNOBYL FALL OUT IN SALT FROM CIECHOCINEK, POLAND

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Abstract The initial interest in the total inventory of the radionuclide fission products ejected from Chernobyl has been replaced with concern about the deposition and accumulation in the environment of those isotopes with longer half lives, mainly Cs-137 and Cs-134. We present here the slightly unusual case of collection of caesium isotopes in the salt of graduation towers used in salt production. In this case, the radiocaesium first settled down from the air to the wetted surface of the graduation tower in Ciechocinek and then, due to crystallisation of the brine, was deposited in the salt.

1. INTRODUCTION

Ciechocinek is situated on the left bank of the Vistula River, not far from Torun. It is a small and quiet town and is one of the leading Polish health resorts, famous for its natural saline waters. The natural springs of brine there have amazed people for hundreds of years. By the 13th Century salt was being manufactured from the brine springs. After the first partition of Poland in 1772 the country lost its salt mines (Wieliczka and Bochnia). Because of this attention was turned to the brine springs in Ciechocinek. Taking up the idea of the Polish scientist and statesman, Stanislaw Staszic, in 1824 the government decided to build graduation towers and salt works for the concentration and crystallisation of brine.

The three graduation towers in Ciechocinek are huge structures (16 m high and 1742 m in total length) built of thick beams supported by wooden stays sunk deep into the ground. These wooden frames are filled with horizontal layers of blackthorn branches (see Fig. 1). The brine percolates down the tower walls and then flows through a pipe to the salt works. The aerosol formed around the tower forms a sea-like microclimate containing natural iodine and bromine anions and is used in a variety of inhalation therapies.

The technology of the salt production has remained basically the same for the past 100 y. Nowadays, the brine from the artesian well is pumped to the top of the graduation tower. This well (No. 11) is 405 m deep and draws the water from a Jurassic aquifer (Dogger water). The salt of brines in this Mesozoic formation ($> 10^8$ y) probably has its genesis in both marine and rain paleoinfiltration waters (Dowgiała, 1971). The blackthorn branches filling the walls of graduation works are gradually but constantly renewed, The average time of replacement is greater than 10 y.

The nuclear accident at the power station in Chernobyl on 26 April 1986 caused radioactive material to be ejected into the atmosphere. The total amount of the radionuclides released from Chernobyl was 2×10^{18} Bq, with an uncertainty of 50%. The most convincing estimate of Cs-137 release is 8.5×10^{16} Bq (Zimina *et al.*, 1993). At the moment of the accident the estimated average ratio of Cs-134/Cs-137 was 0.52-0.54. It should be noted here that, unlike Cs-137, Cs-134 is produced in significant amounts by nuclear reactors but not by nuclear weapon explosions (Shapiro, 1995).

The plume of radioactive pollution from Chernobyl, that affected Europe, initially moved slowly towards the Baltic Region. In Finland and Sweden the activity of 16 identified nuclides was stable ($ca 10$ Bq m^{-3}) during 28-29 April (Devell *et al.*, 1986). Then the cloud moved more quickly over Poland, Alpine territory, France and then England.

2. RESULTS

The distribution of caesium isotopes for the whole of Poland was measured for the first time in October 1988 (Biernacka *et al.*, 1990). The next measurements were during the national programme "Protection of Lithosphere" (Strzelecki *et al.*, 1994), when field work was carried out between June and December 1992. From the map of the distribution of caesium contamination it can be seen that Cs-137 soil contamination increases from the northern to southern parts of Poland. Results indicate that more than 90% of the territory is characterised by low Cs concentration varying from 0 to 8 kBq m^{-2} , Ciechocinek is placed in the region with the lowest caesium concentration (< 3 kBq m^{-2}).

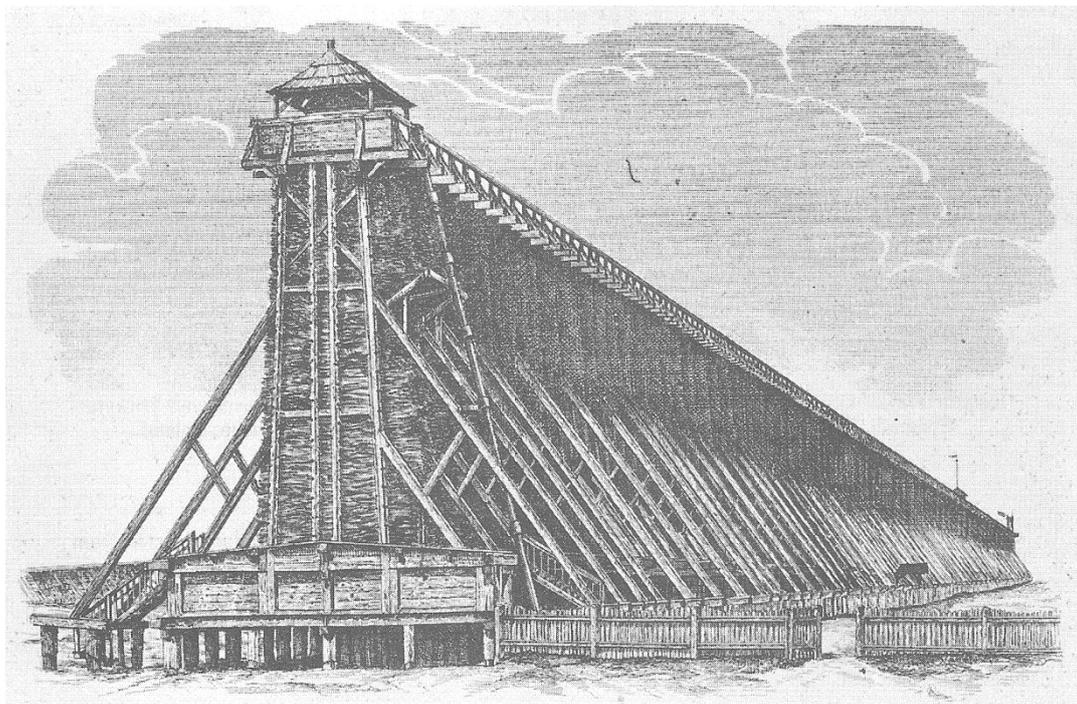


Fig. 1. Intaglio from the old guidebook (L. Mieczkowski, Unger, Warszawa 1873) showing the construction of the graduation tower. The brine percolates down the wooden walls filled with layers of blackthorn branches, increasing the salt concentration.

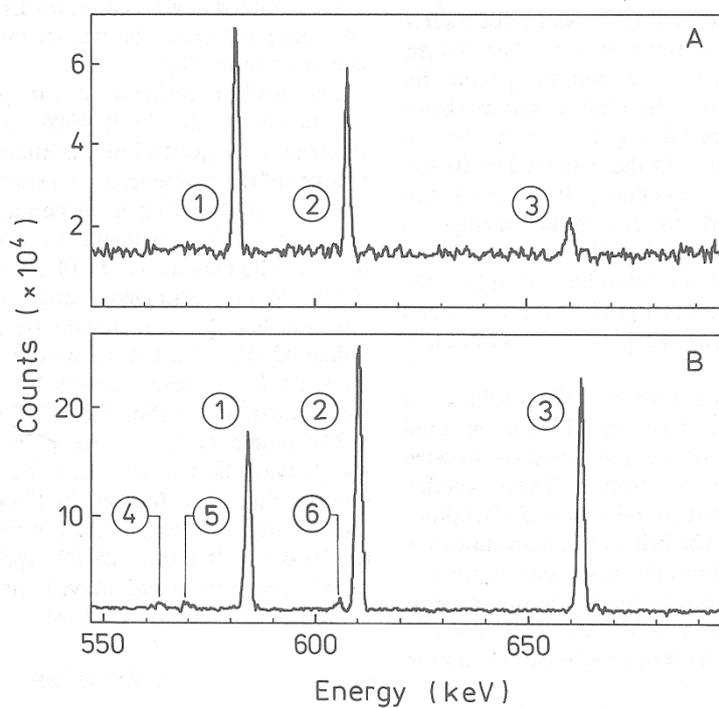


Fig. 2. A selected part of the gamma-ray spectrum of salt from Ciechocinek obtained with the Ge detector (GX 1520) and multichannel analyser Canberra MCA100. The spectrum (A) records the radioactivity of commercial salt (weight: 0.45 kg; time: 168 h; start: 1 September 1995). The labelled lines come from: (1) Tl-208 Th, 583.1 keV; (2) Bi-214 U, 609.3 keV; and (3) Cs-137, 661.6 keV. The spectrum (B) for the crystallised salt removed from the graduation tower (0.75 kg, 96 h, 24 October 1995) shows additional lines from Cs-134; (4) 563.2 keV; (5) 569.3 keV; and (6) 604.7 keV.

Table 1. Radionuclide analysis report. Activity of salt taken directly from graduation tower was measured in October 1995.
The predecay date is taken as 1 May 1986

Nuclide	Sample (Bq/kg)	Predecayed (Bq/kg)	Uncertainty (%)
Cs-137	2.78	3.46	2.60
Cs-134	0.0763	1.85	12.00
K-40	5.35	5.35	4.81
Ac-228 Th	6.29	6.29	1.11
Pb-212 Th	6.26	6.26	2.20
Bi-212 Th	7.32	7.32	3.25
Tl-208 Th	1.91	1.91	2.42
Bi-214 U	6.33	6.33	1.33

Gamma-ray spectrometer--Canberra MCA100 with Sampo90 software provides computerized isotope identification through peak searches of spectra and scans of the nuclide library list. It was a real surprise when the radionuclide analysis of commercial salt from Ciechocinek revealed a small concentration of Cs-137. Its level of activity (0.12 Bq/kg) is substantially lower than that produced by naturally occurring radioactive isotopes.

A relevant portion of the gamma-ray spectrum is shown in Fig. 2(A) and indicates two lines corresponding to the series of natural radioactivity (Tl-208 from the thorium series, 583.1 keV, Bi-214 from the uranium 238 series, 609.3 keV) and the third weak line at 661.6 keV undoubtedly belonging to Cs-137. The half life of Cs-137 (30.2 y) is very short on a geological time scale and can only be from man-made sources and its concentration in salt records artificial contamination.

Considering the technology of salt production in Ciechocinek, the most probable source of salt contamination was the air pollution around the graduation towers. It appears that the graduation towers with their large size and wet walls became an efficient trap for Chernobyl fallout. In order to check this likelihood the gamma-ray spectrum of crystallized salt removed from the blackthorn branches of the tower was measured. The sample was taken from a part of the tower where the blackthorn had not been renewed since 1985. The relevant spectrum is shown in Fig. 2(B) and the radionuclide analysis in Table 1. The predecay date is taken as 1 May 1986.

The radioactivity of salt from the graduation tower is higher than that of the commercial salt and as a consequence it was possible to identify the second artificial isotope δ Cs-134 (half-life 2.07 y) via the spectral lines: 563.2, 569.3, 604.7 and 795.8 keV. It is then possible to calculate the activity ratio Cs-134/Cs-137 at the predecay time of the Chernobyl accident. The calculated ratio of 0.53 derived from our measurements, with an estimated uncertainty of 12% is close to the value which has been estimated for Chernobyl pollution (Devell *et al.*, 1986; Zimina *et al.*, 1993) and confirms the suggested mechanism for salt contamination.

While the presence of Cs-137 and Cs-134 are clearly detectable its level of activity is substantially below that produced by naturally occurring radioactive isotopes.

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