INDUSTRIAL SAFETY

What caused a plume of radioactive ruthenium in Europe in 2017?

Chemical detective work suggests an accident at a Russian plant that processes spent nuclear fuel was the source of the cloud

by ***Laura Howes***

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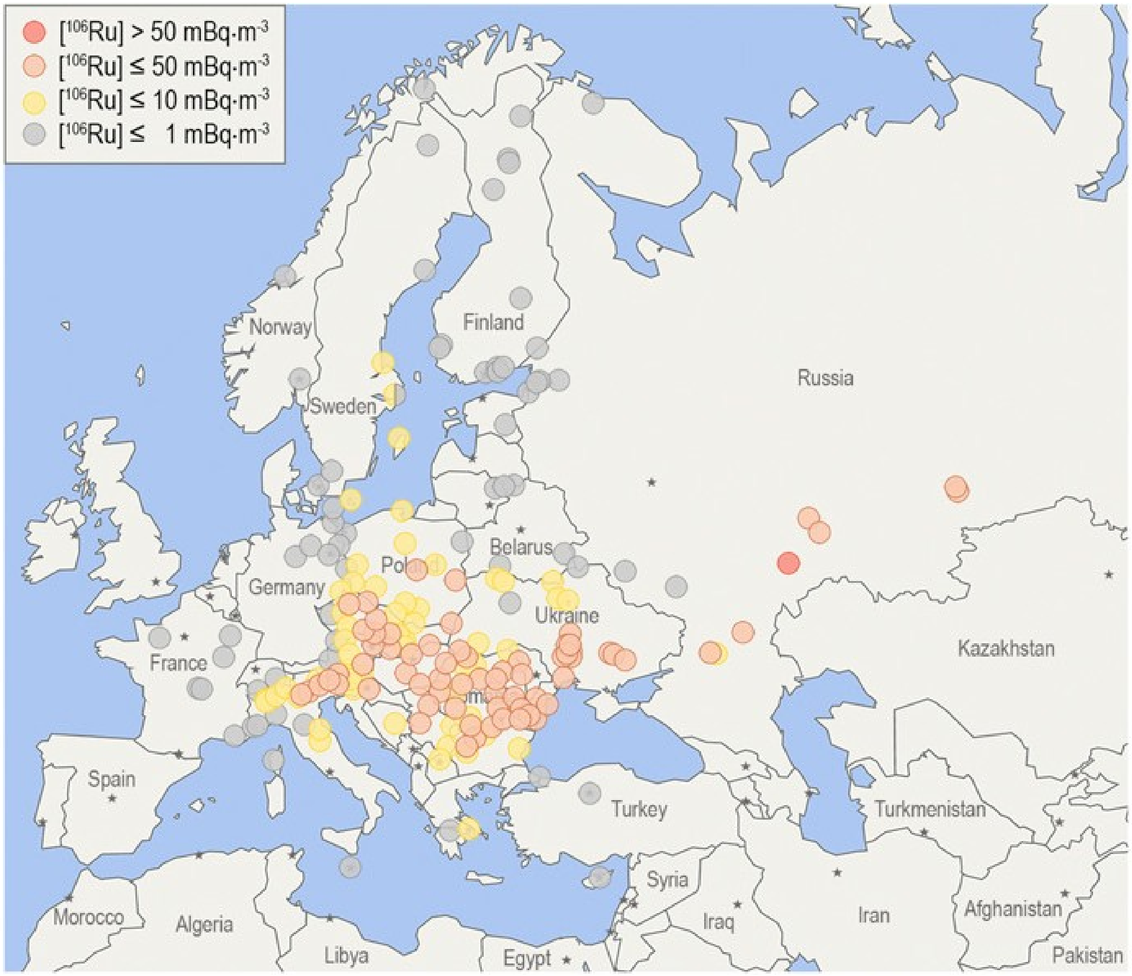

Map of corrected average concentrations of ruthenium-106 at European stations in October 2017. An adult exposed to ruthenium-106 at a concentration of 1 Bq/m^3 for a whole year would only get half the recommended dose limit for that year.

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What caused a plume of radioactive ruthenium in Europe in 2017?

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On an otherwise ordinary Monday in early October 2017, nuclear monitoring sites across Europe started to detect radioactive ruthenium-106 in the filters that they use to collect air samples. Scientists at the labs spotted the isotope when they came to work after the weekend and performed their weekly filter exchange. Members of the informal network between the laboratories, known as the Ring of Five after its founding scientists from five different countries, started calling each other to see if they had recorded similar measurements. When they realized they all were observing the radioactive material, they knew some sort of nuclear event had happened.

The radioactive material never posed a threat to human health because the concentrations were low. But through multiple pieces of evidence, the researchers have concluded that the most likely source for the plume of radioactive ruthenium that traveled across Europe in late September and early October 2017 was a fire or explosion at the Mayak production plant in the Ural Mountains of

Russia sometime between the evening of Sep. 25 and the afternoon of the next day (*Proc. Natl. Acad. Sci USA* 2019 DOI: **10.1073/pnas.1907571116**). This plant was probably processing spent nuclear fuel to create enriched cesium for particle physics experiments at the Gran Sasso National Laboratory in Italy.

"This is an outstanding case study in radiochemical forensics," says Neil Hyatt, an expert in nuclear materials chemistry at the University of Sheffield. The work, he says, "demonstrates the need to maintain high caliber national capabilities, and international partnerships, to support radiological detection and investigation."

When the researchers looked back at data from multiple sites, they determined that the nuclear event happened in late September. They also could pinpoint the possible source of the ruthenium plume by studying the readings from their labs and working back in time to trace it to the Urals. The Russian state weather service, Rosgidromet, also released a statement about detecting **ruthenium-106 at meteorological stations in the southern Ural Mountains**. But the Russian nuclear corporation, Rosatom, denied that there had been any leak or accident at any of its nuclear sites. Russian authorities instead suggested that the ruthenium could have come from the battery of a satellite burning up on reentry.

To get to the bottom of the puzzle, Georg Steinhauser at the University of Hannover in Germany put out a call for the air-sampling filters from the various monitoring sites. His aim was to try and piece together chemically where the ruthenium came from and how it came to be blowing across Europe.

First, Steinhauser and his team established the age of the nuclear fuel by measuring isotopic ratios. The result, he says, was "stunning." When you take nuclear fuel from a reactor, he explains, its radiation levels are enormous and so workers have to wait several years before processing the material. For example, Steinhauser says, the French wait at least 4 years before they even consider reprocessing and the Russians normally wait at least 3 years. But the ruthenium-103-to-ruthenium-^{106} ratios that Steinhauser's team found suggested that the processed fuel would have been very young—only 1.5 to 2 years old. "It was so young, that we actually doubted the quality of our data at first," he admits. "No facility in the world would ever dare to reprocess fuel that is so young." Except, he says, he thinks someone did.

The isotopic ratios, as well as the magnitude of the release, and measurements from meteorological stations at different altitudes, all ruled out the possibility that the ruthenium came from a satellite burning up on reentry, as the Russians had suggested. Instead, Steinhauser says, the data point to the reprocessing of very young nuclear fuel, perhaps to create compact sources of cerium-144 for the use in sterile neutrino experiments at the Gran Sasso lab in Italy. In such experiments, researchers would look for neutrinos by producing antineutrinos that interact with the elusive particles. Cerium-144 provides a source of antineutrinos. A report on the Gran Sasso experiment known as SOX (*J. Phys.: Conf. Ser.* 2016, DOI: **10.1088/1742-6596/675/1/012032TI**) indicates that the Mayak plant took an order for cerium-144 from Gran Sasso in 2016. SOX researchers later reported that the plant canceled the order in December 2017.

A plant could get more cerium-144 by reprocessing young spent fuel but the process would be trickier because of the fuel's high radioactivity. After analyzing the ruthenium particles in the filters, Steinhauser concluded that an accident might have happened while processing the young fuel. High levels of ionizing radiation can affect the chemical reactions involved in the fuel processing and extra heat generated by the spent fuel can warm explosive gases.

To process spent nuclear fuel, workers normally use a method known as PUREX, which chops up the fuel into smaller fragments and then dissolves it in nitric acid. Most of the fission products, Steinhauser says, get dissolved in the aqueous solution, but some ruthenium can get oxidized to

RuO_4 gas, which is usually separated out and processed. The gas can easily escape into the atmosphere through a small leak. But when Steinhauser's group did solubility and volatility tests on the ruthenium trapped in the air filters from the European labs, they found at least two different ruthenium compounds with very different properties. A mix of ruthenium compounds suggested to the researchers that a small gas leak wasn't the source. Instead, Steinhauser says, a fire or explosion at the plant during ruthenium processing could have sent a mix of compounds into the air.

Rosatom, which oversees Mayak, continues to deny accidents at any of their operated plants or facilities. "Both the national regulator and experts from an independent international inquiry inspected the Mayak facility back in 2017 and found nothing to suggest that the ruthenium-106 isotope originated from this site, nor found any traces of an alleged accident, nor found any evidence of local staff exposure to elevated levels of radioactivity," according to a statement from Rosatom emailed to C&EN.

The results of Steinhauser's analysis, which combines different pieces of evidence, is fascinating and seems sound, says Ivana Nikolic Hughes, director of the K=1 Project at Columbia University's Center for Nuclear Issues.

"It is just a hypothesis," Steinhauser admits. "But I would almost bet a lot of money on this is true." The Russians, he says, are experts in processing spent nuclear fuel and so he is unsure why they have not yet come forward with details of what happened. He suspects the reason for their silence may be related to the fact that Mayak is involved in military activity. "We have not fully understood yet about what the implications are," he says. "But the chemistry of the release can tell us something about the happenings in the facility and about the circumstances of the accident."

UPDATE

This story was updated July 31, 2019, to include a statement from Rosatom.

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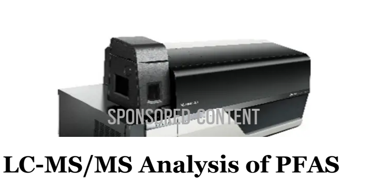

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