

Airborne concentrations and chemical considerations of radioactive ruthenium from an undeclared major nuclear release in 2017

O. Masson^{a,1,2}, G. Steinhauser^{b,1,2}, D. Zok^b, O. Saunier^c, H. Angelov^d, D. Babić^e, V. Bečková^f, J. Bieringer^g, M. Bruggeman^h, C. I. Burbidgeⁱ, S. Conilⁱ, A. Dalheimer^k, L.-E. De Geer^{l,3}, A. de Vismes Ott^m, K. Eleftheriadisⁿ, S. Estier^o, H. Fischer^p, M. G. Garavaglia^q, C. Gasco Leonarte^r, K. Gorzkiewicz^s, D. Hainz^t, I. Hoffman^u, M. Hýža^f, K. Isajenko^v, T. Karhunen^w, J. Kastlander^l, C. Katzberger^x, R. Kierepko^s, G.-J. Knetsch^y, J. Kövendiné Kónyi^z, M. Lecomte^{aa}, J. W. Mieltsch^q, P. Min^{bb}, B. Møller^{cc}, S. P. Nielsen^{dd}, J. Nikolic^{ee}, L. Nikolovska^{ff}, I. Penev^d, B. Petrinec^e, P. P. Povinec^{gg}, R. Querfeld^b, O. Raimondi^{hh}, D. Ransbyⁱⁱ, W. Ringer^{jj}, O. Romanenko^{kk}, R. Rusconi^{ll}, P. R. J. Saey^t, V. Samsonov^{mm}, B. Šilobritienėⁿⁿ, E. Simion^{oo}, C. Söderström^l, M. Šoštarić^e, T. Steinkopf^{kk}, P. Steinmann^o, I. Šykora^{gg}, L. Tabachnyi^{pp}, D. Todorovic^{ee}, E. Tomankiewicz^s, J. Tschiersch^{qq}, R. Tšibranski^{rr}, M. Tzortzis^{ss}, K. Ungar^u, A. Vidic^{tt}, A. Weller^b, H. Wershofen^{uu}, P. Zagyvai^{vv}, T. Zalewska^{ww}, D. Zapata García^{uu}, and B. Zorko^{xx}

^aDirection de l'Environnement, Institut de Radioprotection et de Sûreté Nucléaire, 13115 St Paul lez Durance, France; ^bInstitute of Radioecology and Radiation Protection, Leibniz Universität Hannover, 30419 Hannover, Germany; ^cService des Situations d'Urgence et d'Organisation de Crise, Institut de Radioprotection et de Sûreté Nucléaire, 92260 Fontenay-Aux-Roses, France; ^dBasic Environmental Observatory Moussala, Institute for Nuclear Research and Nuclear Energy, BG-1784 Sofia, Bulgaria; ^eInstitute for Medical Research and Occupational Health, 10001 Zagreb, Croatia; ^fSection of Monitoring, National Radiation Protection Institute, 140 00 Prague 4, Czech Republic; ^gAtmosphärische Radioaktivität und Spurenanalyse, Bundesamt für Strahlenschutz, 79098 Freiburg im Breisgau, Germany; ^hStudieCentrum voor Kernenergie-Centre d'Etude de l'Energie Nucléaire, 2400 Mol, Belgium; ⁱOffice of Radiation Protection and Environmental Monitoring, Environmental Protection Agency, Dublin, D14 H424, Ireland; ^jObservatoire Pérenne de l'Environnement, Agence Nationale des Déchets Radioactifs, 55290 Bure, France; ^kDeutscher Wetterdienst, 63067 Offenbach, Germany; ^lChemical, Biological, Radiological and Nuclear Defence and Security Division, Swedish Defence Research Agency, 16490 Stockholm, Sweden; ^mLaboratoire de Mesure de la Radioactivité dans l'Environnement, Institut de Radioprotection et de Sûreté, 91400 Orsay, France; ⁿInstitute of Nuclear and Radiological Sciences & Technology, Energy & Safety, National Centre for Scientific Research "Demokritos", 15310 Athens, Greece; ^oEnvironmental Radioactivity Section, Federal Office of Public Health, CH-3097 Liebefeld, Switzerland; ^pRadioactivity Measurements Laboratory, University of Bremen, 28359 Bremen, Germany; ^qDipartimento Prevenzione, Agenzia Regionale per la Protezione dell'Ambiente del Friuli Venezia Giulia, 33100 Udine, Italy; ^rUnidad de Radiactividad Ambiental y Vigilancia Radiológica, Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas, 28040 Madrid, Spain; ^sDepartment of Nuclear Physical Chemistry, The Henryk Niewodniczanski Institute of Nuclear Physics, 31-342 Kraków, Poland; ^tAtominstytut, Vienna University of Technology, 1020 Vienna, Austria; ^uHealth Canada, Ottawa, K1A 1C1 ON, Canada; ^vCentral Laboratory for Radiological Protection, PL 03-134 Warsaw, Poland; ^wEnvironmental Monitoring, Radiation and Nuclear Safety Authority, FI-00881 Helsinki, Finland; ^xDepartment of Radiation Protection and Technical Quality Assurance, Austrian Agency for Health and Food Safety, 1220 Vienna, Austria; ^yCentre for Environmental Safety and Security, National Institute for Public Health and the Environment, NL-3720 BA Bilthoven, The Netherlands; ^zDivision of Environmental and Public Radiohygiene, National Research Institute for Radiobiology and Radiohygiene, H-1221 Budapest, Hungary; ^{aa}Division of la Radioprotection, Ministère de la Santé, L-2120 Luxembourg, Luxembourg; ^{bb}National Commission for Nuclear Activities Control, District 5, 050706 Bucharest, Romania; ^{cc}Emergency Preparedness and Response, Norwegian Radiation and Nuclear Safety Authority, NO-9925 Svanvik, Norway; ^{dd}Center for Nuclear Technologies, Technical University of Denmark (DTU) Risø, DK-4000 Roskilde, Denmark; ^{ee}Department of Radiation and Environmental Protection, Institute for Nuclear Sciences, University of Belgrade, 11351 Belgrade, Serbia; ^{ff}Radiology Laboratories, Institute of Public Health, 1000 Skopje, North Macedonia; ^{gg}Department of Nuclear Physics and Biophysics, Comenius University, 84248 Bratislava, Slovakia; ^{hh}Tutela dell'Ambiente Naturale e Costruito, Dipartimento Prevenzione, 47893 Borgo Maggiore, Republic of San Marino; ⁱⁱComputing Centre, Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research, 27570 Bremerhaven, Germany; ^{jj}Department of Radon and Radioecology, Austrian Agency for Health and Food Safety, 4020 Linz, Austria; ^{kk}Rivne Nuclear Power Plant, National Nuclear Energy Generating Company "Energoatom", 34400 Rivne Oblast, Ukraine; ^{ll}Centro Regionale Radioprotezione, Agenzia Regionale per la Protezione dell'Ambiente della Lombardia, 20124 Milan, Italy; ^{mm}Republican Center of Hydrometeorology, Radioactive Contamination Control and Environmental Monitoring, 220114 Minsk, Belarus; ⁿⁿEnvironmental Protection Agency, 09311 Vilnius, Lithuania; ^{oo}National Reference Laboratory, National Environmental Protection Agency, 060031 Bucharest, Romania; ^{pp}Ukrainian Hydrometeorological Center, 01601 Kyiv, Ukraine; ^{qq}Institute of Radiation Medicine, Helmholtz Zentrum München, 85764 Neuherberg, Germany; ^{rr}Safety Department, Kozloduy Nuclear Power Plant, 3321 Kozloduy, Bulgaria; ^{ss}Radiation Inspection and Control Services, Department of Labour Inspection, CY-1080 Nicosia, Cyprus; ^{tt}Radiation Protection Center, Institute for Public Health, 71000 Sarajevo, Federation of Bosnia and Herzegovina; ^{uu}Environmental Radioactivity Laboratory, Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany; ^{vv}Centre for Energy Research, Hungarian Academy of Sciences, 1121 Budapest, Hungary; ^{ww}Institute of Meteorology and Water Management, National Research Institute, 01673 Warsaw, Poland; and ^{xx}Infrastructural Group for Ionizing Radiation Measurements, Institut "Jožef Stefan", 1000 Ljubljana, Slovenia

Edited by John H. Seinfeld, California Institute of Technology, Pasadena, CA, and approved June 21, 2019 (received for review May 2, 2019)

In October 2017, most European countries reported unique atmospheric detections of aerosol-bound radoruthenium (¹⁰⁶Ru). The range of concentrations varied from some tenths of $\mu\text{Bq}\cdot\text{m}^{-3}$ to more than $150\text{ mBq}\cdot\text{m}^{-3}$. The widespread detection at such considerable (yet innocuous) levels suggested a considerable release. To compare activity reports of airborne ¹⁰⁶Ru with different sampling periods, concentrations were reconstructed based on the most probable plume presence duration at each location. Based on airborne concentration spreading and chemical considerations, it is possible to assume that the release occurred in the Southern Urals region (Russian Federation). The ¹⁰⁶Ru age was estimated to be about 2 years. It exhibited highly soluble and less soluble fractions in aqueous media, high radiopurity (lack of concomitant radionuclides), and volatility between 700 and 1,000 °C, thus suggesting a release at an advanced stage in the reprocessing of nuclear fuel. The amount and isotopic characteristics of the radoruthenium release may indicate a context with the production of a large ¹⁴⁴Ce source for a neutrino experiment.

Author contributions: O.M. and G.S. designed research; O.M., G.S., D.Z., O.S., H.A., D.B., V.B., J.B., M.B., C.I.B., S.C., A.D., L.-E.D.G., A.d.V.O., K.E., S.E., H.F., M.G.G., C.G.L., K.G., D.H., I.H., M.H., K.I., T.K., J.K., C.K., R.K., G.-J.K., J.K.K., M.L., J.W.M., P.M., B.M., S.P.N., J.N., L.N., I.P., B.P., P.P.P., R.Q., O. Raimondi, D.R., W.R., O. Romanenko, R.R., P.R.J.S., V.S., B.Š., E.S., C.S., M.S., T.S., P.S., I.S., L.T., D.T., E.T., J.T., R.T., M.T., K.U., A.V., A.W., H.W., P.Z., T.Z., D.Z.G., and B.Z. performed research; O.M., G.S., O.S., L.-E.D.G., H.F., I.H., J.W.M., P.P.P., P.R.J.S., K.U., and H.W. analyzed data; O.M., G.S., and D.Z. wrote the paper; and G.S. and A.W. acquired funding.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

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¹O.M. and G.S. contributed equally to this work.

²To whom correspondence may be addressed. Email: olivier.masson@irsn.fr or steinhauser@irs.uni-hannover.de.

³Retired.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1907571116/-DCSupplemental.

Published online July 26, 2019.

Significance

A massive atmospheric release of radioactive ^{106}Ru occurred in Eurasia in 2017, which must have been caused by a sizeable, yet undeclared nuclear accident. This work presents the most compelling monitoring dataset of this release, comprising 1,100 atmospheric and 200 deposition data points from the Eurasian region. The data suggest a release from a nuclear reprocessing facility located in the Southern Urals, possibly from the Mayak nuclear complex. A release from a crashed satellite as well as a release on Romanian territory (despite high activity concentrations) can be excluded. The model age of the radioruthenium supports the hypothesis that fuel was reprocessed ≤ 2 years after discharge, possibly for the production of a high-specific activity ^{144}Ce source for a neutrino experiment in Italy.

environmental radioactivity | ruthenium | nuclear forensics | environmental release | accidental release

Nuclear accidents are serious threats due to their immediate and perceived consequence for both health and environment. The lay public thus relies on the responsibility of their leaders to provide information on radioactive releases and their impact on human and environment health. Early in the 1960s, and even more after the Chernobyl accident, European radioprotection authorities established or strengthened radionuclide monitoring networks on a national scale. Today most of these European networks are connected to each other via the informal “Ring of Five” (Ro5) platform for the purpose of rapid exchange of expert information on a laboratory level about airborne radionuclides detected at trace levels. The Ro5 was founded in the mid-1980s by 5 member countries: Sweden, Federal Republic of Germany, Finland, Norway, and Denmark. Today, the memberships have grown to laboratories in 22 countries (while the name was kept), and the Ro5 is still an informal arrangement on a laboratory level and between scientists. In January 2017, the Ro5 alerted its members regarding the widespread detection of airborne ^{131}I in Europe (1). In October 2017, an unprecedented release of ruthenium-106 (^{106}Ru ; $T_{1/2} = 371.8$ d) into the atmosphere was the subject of numerous detections and exchanges within the Ro5. The goal of this report is to give an overview of the global spreading of this fission product through airborne concentrations observed in Europe and beyond, its forensic history, and chemistry.

Chronology of the Event

On October 2, 2017, an informal alert by an Italian laboratory was issued to the Ro5 network, reporting the detection of airborne ^{106}Ru in the millibecquerel per cubic meter ($\text{mBq}\cdot\text{m}^{-3}$) range in Milan, Italy. Limits of detection (LOD) in laboratories connected to the Ro5 are typically in the range of 0.1 to 10 microbecquerels per cubic meter ($\mu\text{Bq}\cdot\text{m}^{-3}$). This first report occurred on a Monday, when most European laboratories usually exchange their aerosol filters, which are operated on a weekly basis. Later that day, ^{106}Ru detections were reported from Czech Republic, Austria, and Norway in the 1- to $10\text{-mBq}\cdot\text{m}^{-3}$ range. This widespread detection in such range immediately suggested a considerable release.

After 2 d (and further detection reports from Poland, Austria, Switzerland, Sweden, and Greece), official information notes were published by national radioprotection authorities, for example, in Switzerland, Austria, and Norway. On October 7, 2017, the International Atomic Energy Agency (IAEA) requested data and possible known sources of radioruthenium from all 43 European member states. On October 9, 2017, Chelyabinsk and Sverdlovsk regional authorities ruled out any possible ^{106}Ru release from their region (Russian Federation). On November 21,

2017, the Russian Federal Service for Hydrometeorology and Environmental Monitoring (Roshydromet) declared to have measured ^{106}Ru in the southern Urals in the late September (2). However, one possible source in the region, the Federal State Unitary Enterprise “Production Association Mayak” in Ozersk, immediately declared that it was not the source of increased ^{106}Ru (3). On November 23, 2017, the IAEA addressed the release of ^{106}Ru in a press conference. All members submitted the requested data, but none declared an accident and none declared being aware of any source. On December 8, 2017, Russian officials once again claimed that Mayak could not be the source because of the lack of any radioruthenium traces in the soil around the facility (4). Instead, the officials pointed at the possibility of a radionuclide battery of a satellite that had burned during its reentry into the atmosphere. On January 22, 2018, the Nuclear Safety Institute of the Russian Academy of Sciences invited radiation protection experts from Germany, France, Finland, Sweden, the United Kingdom, and Russia to aid in the elucidation of the release. Two commissions of inquiry were held: on January 31, 2018 and on April 11, 2018. The second meeting concluded by emphasizing that not enough data were yet available to point out any verified hypothesis of the origin of the ^{106}Ru (5). The present article aims at closing this gap.

Results and Discussion

Monitoring Results. Information sources: The entire airborne concentration dataset and deposition dataset are available as *SI Appendix, Tables S1–S4* and were mainly compiled through Ro5 exchanges, personal exchange, and data already published. Valuable information are also available on the Roshydromet website (6, 7), on the Typhoon Association website (6), and on the website of the Unified State Automated Monitoring System of the Radiation Situation in the Russian Federation (8). Data from the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) (9) are not part of the dataset except for those already published (10). The International Monitoring System (IMS) data supporting the CTBTO (9) are available directly from the CTBTO upon request and signing a confidentiality agreement to access the virtual Data Exploitation Platform. This study provides ^{106}Ru observations ($>1,120$ data points related to airborne activity and about 200 data points for deposited contamination, from about 330 sampling locations) that can be used for the purpose of atmospheric dispersion and deposition model validation.

With the exception of the westernmost parts of Europe, most monitoring stations reported detections of ^{106}Ru in the range between some tenths $\mu\text{Bq}\cdot\text{m}^{-3}$ meter and more than a $100\text{ mBq}\cdot\text{m}^{-3}$. Fig. 1 illustrates the maximum activity levels per country. Activity concentrations in the millibecquerel per cubic meter range were reported between September 29, 2017 and October 7, 2017, exhibiting a short build-up and rapid decline behavior. The last traces of the plume (microbecquerel per cubic meter range) were measured in sampling periods ending between October 12, 2017 and the end of October by laboratories equipped with high-volume samplers and low-level γ -ray spectrometry. The eastern and southeastern parts of Europe, including western Russia, exhibited the highest reported levels. The maximum level in Europe was reported from Romania ($176 \pm 18\text{ mBq}\cdot\text{m}^{-3}$). Even at this level, the plume did not represent any threat for human and environmental health.

However, it is important to note that these compiled data were obtained with different sampling durations, which limits their comparability without further correction.

Outside Europe, ^{106}Ru was also detected east of the geographical border between Europe and Asia in the Urals region (Russian Federation) with activity levels of some tens $\text{mBq}\cdot\text{m}^{-3}$. Tiny amounts of ^{106}Ru were also pointed out elsewhere in the northern hemisphere by aerosol stations belonging to the IMS supporting the CTBTO: in Guadeloupe, Kuwait, Florida (United States), Russia (central and eastern parts), and Mongolia. ^{106}Ru is a

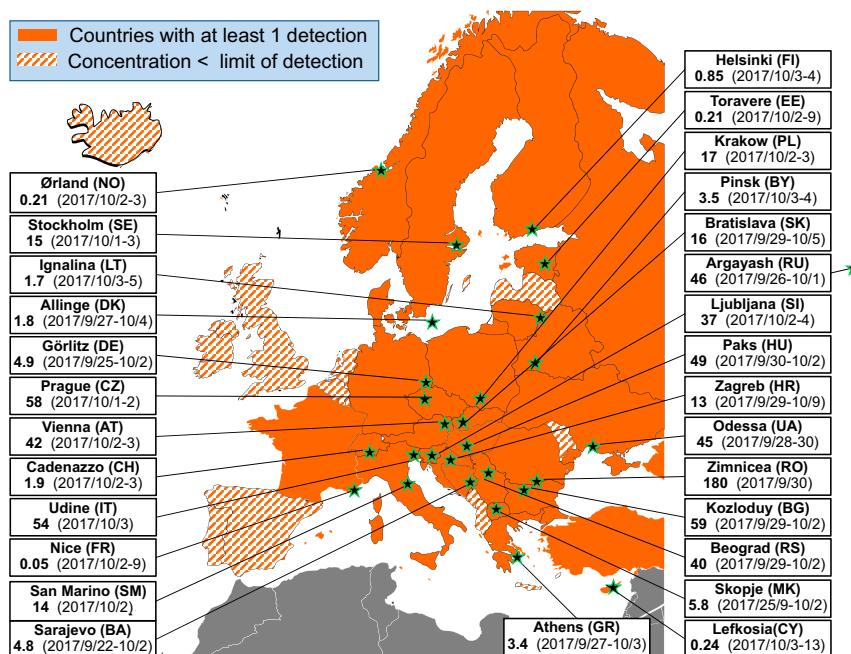


Fig. 1. Compiled maximum ¹⁰⁶Ru airborne “uncorrected” activity concentrations (in mBq·m⁻³; sampling period in parentheses) in Europe.

nuclide that may be released upon detonation of a nuclear weapon, and is therefore a “CTBT-relevant radionuclide.”

¹⁰⁶Ru had not been detected in the global atmosphere since the Chernobyl accident (11) [estimated release <73 PBq (12)], not even after the Fukushima accident on the Japanese territory (13, 14), because of the different accident and release characteristics. As a result, there is no usual background or reference level, which could be used to define an increasing factor. As a matter of fact, this radionuclide is usually not detected in the atmosphere. Besides aerosol filtration, gaseous sampling was conducted at some locations (Austria, Sweden, Italy, and Poland), thus allowing checking for the presence of gaseous Ru species. Ruthenium may be present in volatile forms, especially in the form of ruthenium tetroxide, RuO₄ (15). Since gaseous RuO₄ is a highly reactive and strong oxidizer, it is expected to rapidly nucleate into particulate and low volatile RuO₂. No ¹⁰⁶Ru was detected in gaseous form.

In addition to ¹⁰⁶Ru, the anthropogenic ruthenium isotope ¹⁰³Ru ($T_{1/2} = 39.3$ d) was detected at a limited number of high-performing stations (*SI Appendix, Table S2*): Austria, Czech Republic, Poland, and Sweden (10) with activity levels ranging from 0.04 to 7.3 μBq·m⁻³ (average 2.6 ± 0.1 μBq·m⁻³). The average ratio ¹⁰³Ru/¹⁰⁶Ru was about $(2.7 \pm 0.9) \cdot 10^{-4}$, and the minimum ¹⁰⁶Ru activity concentration associated with a ¹⁰³Ru detection was about 4 mBq·m⁻³ (*SI Appendix, Table S4*). Several organizations in Europe analyzed the filters for the occurrence of other γ-emitters, as well as difficult-to-measure radionuclides, such as Pu, Am, Cm, or ⁹⁰Sr by low-level

radiochemical analyses. No unusual traces were found that would have been indicative of a release of any of these radionuclides concurrently with the ^{103,106}Ru. This excludes an accidental release from a nuclear reactor as the source, as this would have resulted in an emission of a great multitude of fission products. Instead, the origin of ^{103,106}Ru is rather associated with nuclear fuel reprocessing or with (medical or technical) radioactive sources. In addition, no unusual (stable) element contamination was found on a ¹⁰⁶Ru-containing filter from Vienna (Austria) (16).

Discussion of a Possible Source Melting. Melting of radioactive sources already occurred in the past, leading to detection of radionuclides in several European countries. Indeed, at the end of May 1998, a ¹³⁷Cs source estimated 0.3 to 3 TBq was incidentally melted in a steelworks near Algeciras (Spain) and led to detections in several European countries (17). ¹⁰⁶Ru in ophthalmic radiotherapy sources have typical activities less than 10 MBq, which is clearly insufficient to explain the observed concentrations on a wide scale, as emphasized by the IAEA (18), as it would have required the melting of numerous ophthalmic sources at once.

Discussion of a Possible Satellite Reentry. The possibility of the disintegration of a satellite equipped with a radioisotope thermoelectric generator (RTG) operated with a ¹⁰⁶Ru source during its reentry into the atmosphere, as vaguely indicated previously (19), warrants investigation. Generally, such a source appears rather

Table 1. Comparison of airborne ¹⁰⁶Ru at high-altitude and at closest low-altitude sampling locations (with similar time stamps)

Country	High-altitude location (meters above sealevel)	Date (2017)	¹⁰⁶ Ru (mBq·m ⁻³)	Nearest low-altitude location	Date (2017)	¹⁰⁶ Ru (mBq·m ⁻³)
Austria	Sonnblick (3,105)	October 2	0.17 ± 0.11	Klagenfurt	October 2	4.90 ± 0.34
Bulgaria	Moussala peak (2,925)	October 3	1.8 ± 0.54	Yana	September 29 to October 4	17.6 ± 1.6
Greece	Helmos Mountain peak (2,314)	September 27 to September 29	0.99 ± 0.19	Athens	September 27 to October 3	2.64 ± 0.63
Germany	Zugspitze (2,964)	September 25 to October 2	<0.026	Garmisch-Partenkirchen	September 25 to October 2	<0.023

unlikely because of the rather short half-life of ^{106}Ru compared with the expected or desired satellite life span and the low power level ($\sim 33 \text{ W}\cdot\text{g}^{-1}$) generated by a ^{106}Ru -powered RTG and radiation protection issues during its manufacture and handling. In addition, several space organizations concluded that no satellite went missing during the ^{106}Ru episode (*SI Appendix*). Other arguments are also not in favor of the satellite disintegration hypothesis. If a satellite had burned during its reentry into the atmosphere, it would have caused a vertical distribution of ^{106}Ru in the air: the higher the altitude, the higher the concentration. However, ^{106}Ru at high-altitude locations was either below LOD or significantly lower than ^{106}Ru registered above LOD at low altitude (Table 1). The very low level (below LOD) at the station on Zugspitze mountain (Germany) is also very indicative of low concentrations at high altitude. In addition, the levels of ^7Be (a cosmogenic radionuclide produced in the upper troposphere/lower stratosphere and used as a tracer of atmospheric movement) remained close to usual range, thus indicating that no downdraft from the lower stratosphere or upper troposphere occurred at that time. Therefore, the ^{106}Ru release has likely happened in the lower tropospheric layers and cannot be linked to a satellite disintegration. Moreover, the concomitant detections of infinitesimal traces of ^{103}Ru and traces of ^{106}Ru at some locations definitely outmoded the satellite reentry hypothesis due to the short ^{103}Ru half-life ($T_{1/2} = 39.3 \text{ d}$).

Sequence of Airborne ^{106}Ru Detections in Europe. Most early ^{106}Ru detections occurred on the aerosol filters sampled during week 39 (September 25 to October 2, 2017), regardless of the location in Europe, including Russia (September 26 to October 1, 2017). In Ukraine, ^{106}Ru was first detected in the sampling period from September 22 to September 29, 2017. Within the framework of national monitoring programs of airborne radionuclides, most aerosol samplings in Europe are performed either on a weekly basis for γ -counting or on a daily basis (in the vicinity of nuclear facilities), first for the purpose of gross β -counting by plant operators, or for γ -counting, for example, within the IMS network. Aerosol filters sampled in Romania were acquired with the highest temporal resolution (down to 5 h plus 1-h shutdown intervals). After sampling, filters of 2 or 4 sequences were compiled before measurement, thus reflecting 10-h sampling of 12 to 20 h of 24 for each composite sample. In addition, the Romanian network consists of

several tens of aerosol sampling stations, which made it possible to reconstruct the pattern of the ^{106}Ru plume. The duration of the episode proved to be rather short: at more than 30 Romanian sampling locations that detected ^{106}Ru , this radionuclide was detectable on exclusively 1 d (30% of sampling locations), on 2 consecutive days (45%) and on 3 consecutive days (25%). Detections over 2 or 3 consecutive days indicate that the plume presence in Romania was rather short and characterized by a narrow peak (Fig. 2).

The shapes of the Romanian time series match with a short release (i.e., typically less than 1 d), subject to the wind direction did not vary a lot during transport and that the plume border did not undulate while passing at the sampling locations. The detection pattern also provides vague distance-related information on the release point of ^{106}Ru , as multiple Romanian stations detected the plume simultaneously. This is only possible if the plume originated at a sufficiently remote release point to have time to widen to the width of Romania (approximately 600 km) (Fig. 3). Although highest activity concentrations of this ^{106}Ru episode ($>100 \text{ mBq}\cdot\text{m}^{-3}$) have been reported for Romania, the width of the plume supports excluding a release point on the Romanian territory.

All eastern Romanian stations reported airborne ^{106}Ru on September 29, 2017. From September 30, 2017, the general trend indicated a ^{106}Ru front traveling westwards. Peak values were noticed between September 29 and October 1, 2017, depending on the location. On October 1, 2017, eastern stations ceased their detections. From October 4, 2017, no more detection was reported from Romania. In Bulgaria, the ^{106}Ru plume was also assumed to be present only 3 to 4 d (mostly from October 2 to October 4, 2017) (20), about 3 d in Austria and Czech Republic, and over 4 d in Hungary (21). These observations clearly confirm both the shortness of the plume length and the eastern origin of the plume. The discussion of the plume duration exemplifies that in many cases, the sampling duration was longer than the plume duration (21). As a result, a significant fraction of uncontaminated air was pumped through the filter, thus “diluting” the ^{106}Ru activity concentration in most cases. To encompass the entire plume duration regardless of the location, we chose a 7-d integration period. In practice, this decreases the average airborne ^{106}Ru concentration for locations where the ^{106}Ru plume was detected over a period

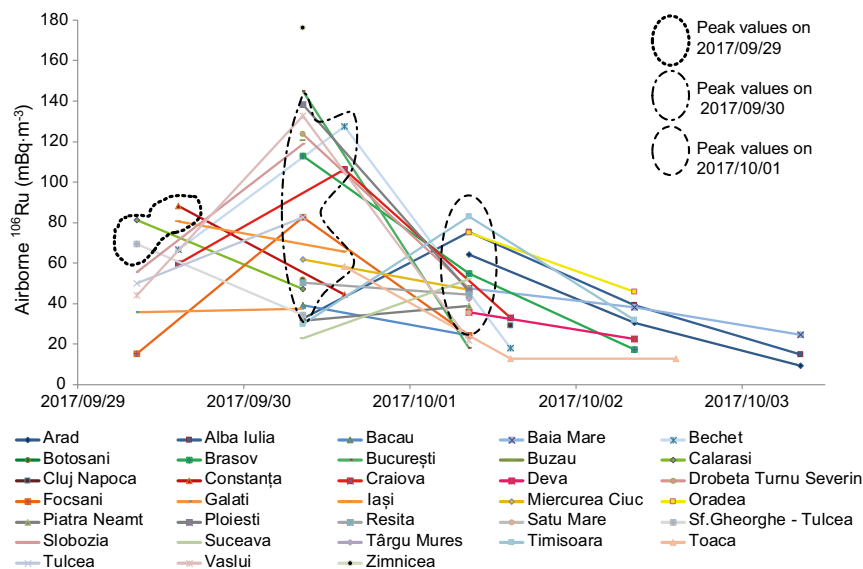


Fig. 2. Airborne ^{106}Ru concentrations ($\text{mBq}\cdot\text{m}^{-3}$) at Romanian locations (values have been attributed to the midsampling date of the composite samples). The connecting lines between data points are only meant to guide the eye.

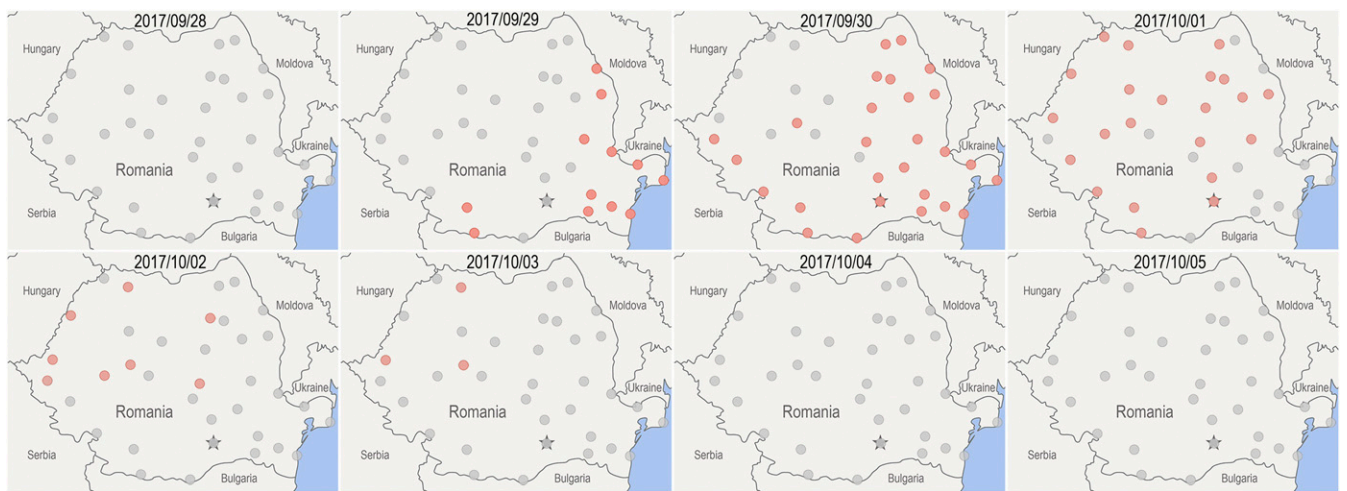


Fig. 3. Daily maps of above-LOD airborne ^{106}Ru (red dots) in Romania from September 28 to October 5, 2017. Gray dots indicate sampling locations with ^{106}Ru levels below the respective limits of detection at the given time.

of <7 d (because the absolute amount of ^{106}Ru was mathematically “diluted” with more clean air), while it consequently increases the ^{106}Ru concentration for sampling periods >7 d (as the plume is mathematically “concentrated” with a smaller amount of air) (Fig. 4). This mathematical unification of the sampling periods lets that the corrected values obtained in Romania no longer stand out as the highest, while it can be observed that they are in the same range from the Urals to southern-central Europe as a result of the conservation of the absolute amount of ^{106}Ru transported along the route of the air masses.

At the Romanian laboratory of Zimnicea—that is, the location with the highest uncorrected value ($176 \pm 18 \text{ mBq}\cdot\text{m}^{-3}$, detected on September 30, 2017 between 3 AM and 2 PM local time)—a Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) back-trajectory analysis shows that air masses came from Russia and then traveled across Ukraine (Fig. 5).

The trajectory model suggests that air masses arriving at Zimnicea on September 30, 2017 had previously traveled close to

the Mayak industrial complex around September 25, 2017 at an altitude of at most about 500 m. According to Roshydromet (2), the meteorological situation in the Southern Urals and central part of European Russia during the period of September 25 to October 6, 2017 was due to a vast anticyclone centered around the White Sea (south of the Kola Peninsula) practically merging with an anticyclone in the central part of Western Siberia. Their report (2) reads, “As a result, conditions for an active transfer of air masses and pollutants from the territory of the Southern Urals and southern Siberia to the Mediterranean region and, then, to the north of Europe, arose in the southern part of Western Siberia, in the Southern Urals, in the Caspian lowland and Ciscaucasia.” Among the different every 3-h back-trajectories, only 2 passed very close to the Mayak industrial complex. The detection of the ^{106}Ru plume at Zimnicea on September 30, 2017 hence indicates a release from Mayak would have occurred between September 25, 2017, around 6 PM coordinated universal time (UTC) and September 26, 2017, around noon (UTC) (Fig. 5).

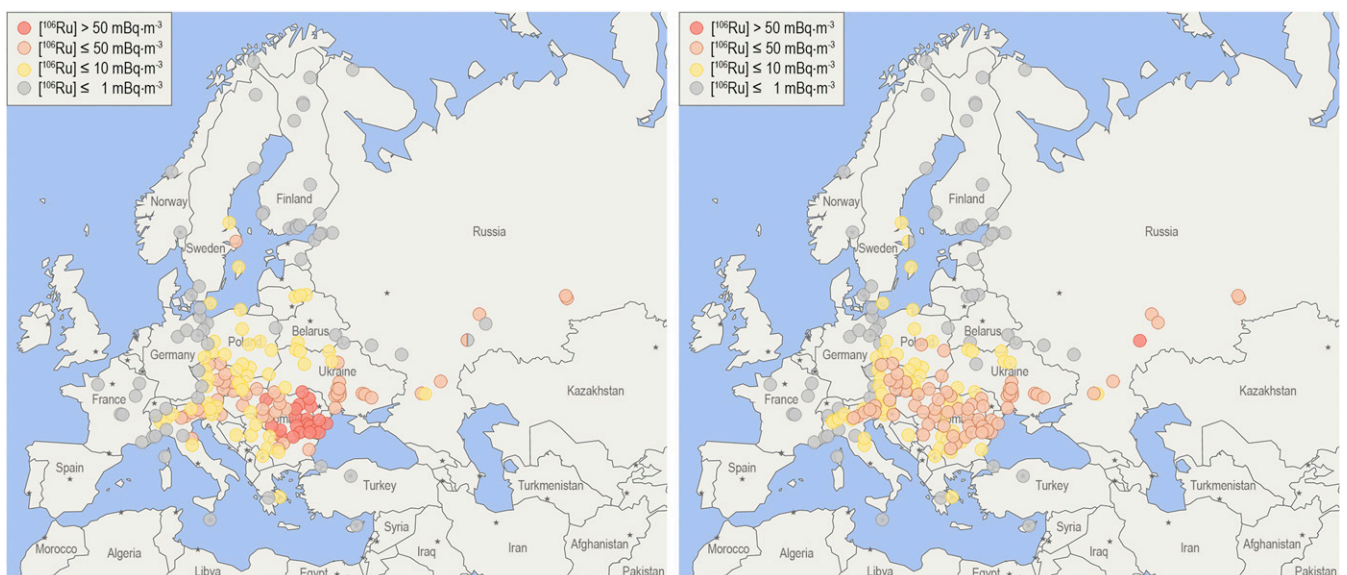


Fig. 4. (Left) Map of uncorrected average concentrations at European stations, and (Right) map of 7-d corrected average concentrations (based on average plume duration of 7 d at each location).

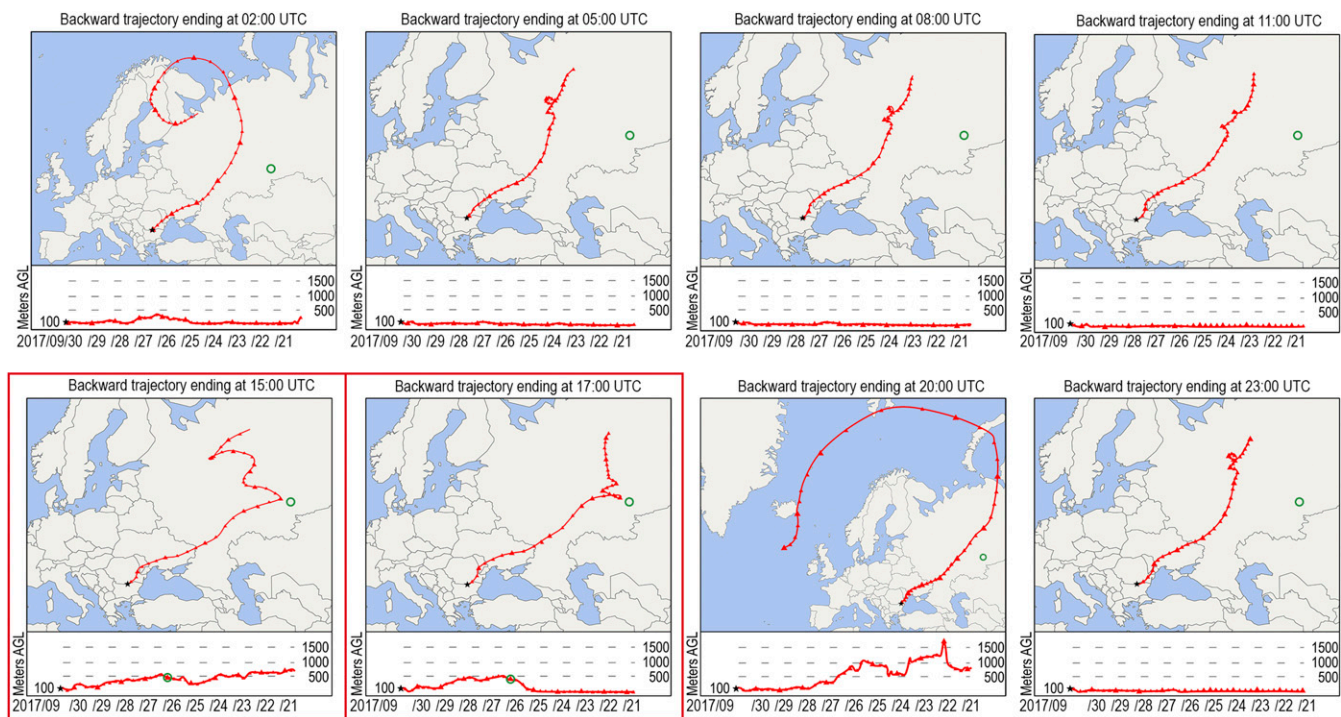


Fig. 5. HYSPLIT-based 240-h backward trajectories ending at the Romanian monitoring station in Zimnicea (black star) (43.666 N, 25.666 E), every 3 h on September 30, 2017, from 2 AM to 11 PM UTC. The National Oceanic and Atmospheric Administration (NOAA) HYSPLIT model uses Global Data Assimilation System (GDAS) meteorological data. The Model Vertical Velocity was used as vertical motion calculation method. The green circle indicates the position of the Mayak industrial complex. The altitude of the air parcels is given in meters above ground level (AGL). The green circle in the altitude sections of the trajectories ending at 3 PM and 5 PM UTC (maps surrounded by red frames), respectively, indicates the time and altitude (approximately 500 m) the air parcels were in closest proximity to the Mayak area.

The Mayak Production Association was one of the first and largest nuclear facilities of the former Soviet Union and spearheaded the Soviet nuclear weapons program. In the Cold War era, it hosted a total of 10 nuclear reactors, mainly for the production of weapons-grade plutonium. In 2014, the Mayak complex employed ~12,000 people and hosted 2 reactors for isotope production, storage facilities for nuclear materials, and a nuclear fuel reprocessing facility (22). On September 29, 1957, a chemical explosion took place in a radioactive waste storage tank at the Mayak nuclear complex, causing a massive release of radionuclides. The accident became known as the “Kyshtym accident.” In the course of this accident, about 2,700 TBq of ^{106}Ru (together with various other radionuclides) were released into the environment, causing a significant contamination in a more than 100-km-long strip that has been termed the East Urals Radioactive Trace (23). More than 10,000 residents had to be evacuated (24). The Kyshtym accident was retrospectively rated at level 6 on the International Nuclear and Radiological Event Scale.

Although such incidents have become rare events in recent years, ^{106}Ru was released from nuclear reprocessing facilities in the past on multiple occasions. On September 26, 1973, following an exothermic reaction at the Windscale reprocessing plant (United Kingdom), 35 workers were contaminated through an atmospheric release of ^{106}Ru estimated at 0.37 TBq (25). On April 6, 1993, an explosion at the reprocessing plant of the Toms-7 nuclear complex (Siberia, Russian Federation) led to the release of approximately 0.52 TBq of ^{106}Ru among other fission products and actinides (26, 27). About 200 km² were contaminated. On May 18, 2001 and October 31, 2001, a failure in the vitrification shops at the La Hague reprocessing plant (France) led to an atmospheric release of ^{106}Ru . Based on aerosols sampled at 200 km downwind from the stack and grass sampled in the vicinity, the first release was estimated between 0.005 and 0.05 TBq, while the

second was estimated to range between 0.0005 and 0.02 TBq (28). Significant atmospheric releases also occurred from the early Hanford operations that were linked to United States nuclear weapons production, with ^{106}Ru (14 TBq from 1944 to 1972) being a relatively minor constituent (compared with 2.7 EBq ^{131}I in the same time span) (29). For comparison, the present, undeclared accident released an estimated activity of 250 TBq at once.

Ruthenium Deposition across Europe. Besides airborne activity determinations, several rainwater, plant, and soil samples attested the deposition of ^{106}Ru across Europe (*SI Appendix, Tables S3 and S4*). Most deposition arose from rain events that occurred between the last week of September 2017 and the first week of October, as for example at several Scandinavian sampling locations (up to about 50 Bq·m⁻² in Sweden and about 50–90 Bq·m⁻² in Finland) or, for example, in Greece in the second week of October (30). In Poland (up to about 80 Bq·m⁻²), a washout ratio ($[^{106}\text{Ru}_{\text{rain}}]/[^{106}\text{Ru}_{\text{air}}]$) of at least 4,900 was found. In central Europe, fallout deposition reached 5 Bq·m⁻² in Vienna (Austria) between October 3 and October 5, 2017; 40 Bq·m⁻² in Ostrava (Czech Republic) from October 2 to October 3, 2017; and 8 Bq·m⁻² in Udine (northeastern Italy) the last week of September 2017 and the first week of October (*SI Appendix, Tables S3 and S4*). The majority of the highest surface-deposition records have been reported from locations within 20 km from the Mayak complex: Khudaiberdinskiy, Argayash, Novogorny, and Metlino (31, 32), where the surface deposition reached up to 343 Bq·m⁻². However, the sole accumulation of positive reports from the vicinity of the Mayak facilities, by itself, is not a conclusive indication of the source, as a nuclear facility naturally is more densely monitored than nonnuclear areas. Depending on the official Russian source, levels are highly variable: up to a factor of 10 that can arise from the deposition pattern.

For the purpose of clarifying the situation, a soil-sampling campaign was conducted by the French Commission for Independent Research and Information on Radioactivity (CRIIRAD) non-governmental organization (33) around the Mayak facilities in December 2017 at a closest authorized distance of about 16 km. Among the 8 soils sampled in various directions, only the 1 sampled west-southwest from Mayak indicated a ^{106}Ru deposited activity estimated between 580 and $1,200\text{ Bq}\cdot\text{m}^{-2}$. Inasmuch as abnormal, again, this sole result is not sufficient to clearly demonstrate whether or not the ^{106}Ru originated from Mayak, as deposition levels are not as high as one might expect from a major release. However, the weak soil-sampling density may also be a good reason for the plume deposition to escape from the investigation grid. Moreover, the atmospheric behavior (e.g., the transfer kinetics from volatile RuO_4 into particulate RuO_2) and deposition of ^{106}Ru are not well understood, especially when released in its volatile form of RuO_4 . At a longer distance (530 km in Bugulma, Russian Federation) in the same direction, deposition of ^{106}Ru (noticed only on samples collected on September 26 to September 27, 2017 [$11.3\text{ Bq}\cdot\text{m}^{-2}$] and September 27 to September 28, 2017 [$30\text{ Bq}\cdot\text{m}^{-2}$]) matches with the hypothesis of a release from the Mayak industrial complex on September 25, 2017, which, therefore, has to be considered as a possible candidate for the source of the release of ^{106}Ru . A detailed dispersion analysis using inverse dispersion modeling techniques and field observations using the data from the present study (airborne concentration and deposition) was conducted to assess both the source location and the source term. This modeling work also suggests that a hot-spot of ^{106}Ru deposition occurred in southeastern Bulgaria. Accordingly, pine needles, oak leaves, forest litter, grass, and soil samples from this area confirm that ^{106}Ru deposition originating from a release on September 26, 2017 from the Mayak area was prone to produce these depositions in combination with rain events.* The ^{106}Ru -deposited activity in plants sampled in the southeast area of Bulgaria was up to several tens $\text{Bq}\cdot\text{kg}^{-1}$, whereas they remained in the millibecquerel per kilogram ($\text{mBq}\cdot\text{kg}^{-1}$) range in the western part of the country where no rain occurred, while the variability of ^{106}Ru volume activity in the atmosphere remained somewhat limited across the country. Other ^{106}Ru detections occurred in early 2018 (until March) in fallout and rainwater samples (in Norway, Poland, Slovenia), and even in March 2019 (Poland), but they were assumed to be induced by the resuspension of previously contaminated soil particles, indicating that ^{106}Ru had not yet completely migrated from the topsoil layer.

Radioruthenium Forensics: Age Estimate, Chemistry, and Volatility.

Model age. Concomitant presence of minute amounts of shorter-lived ^{103}Ru together with ^{106}Ru at about 15 locations allowed estimating the model age of fission-derived radioruthenium (Fig. 6). We define the model age as the time elapsed after the end of neutron irradiation of the nuclear fuel. In a simplified approach, this roughly coincides with the discharge (unloading) of the spent nuclear fuel from the reactor. The average $^{103}\text{Ru}/^{106}\text{Ru}$ activity ratio was found to be in the range of $(2.7 \pm 0.9) \cdot 10^{-4}$, suggesting an age of the released radioruthenium between 530 and 590 d after the end of irradiation in a power reactor [assuming spent nuclear fuel at the end of its fuel lifetime, stemming from a standard power reactor, depending on reactor type and fuel (34) and calculating decay according to the nuclides' half-lives].

There have been speculations in a *Science* commentary (35) that the release of ^{106}Ru may have been associated with the production of a powerful cerium-144 (^{144}Ce ; $T_{1/2} = 285\text{ d}$) source at Mayak for a neutrino experiment in the Gran Sasso National

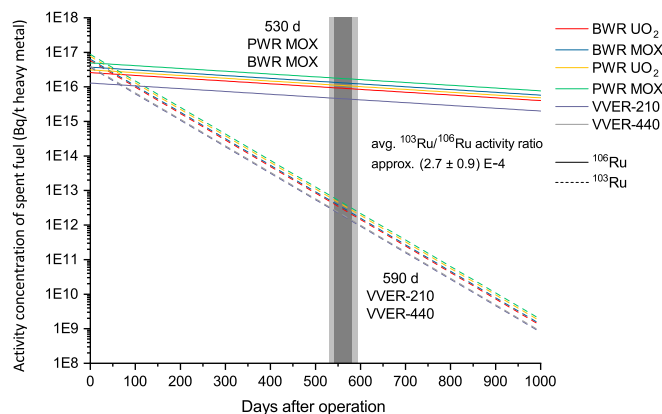


Fig. 6. Age estimation of radioruthenium from various power reactor types (boiling water reactor, BWR; pressurized water reactor, PWR; Russian Water-Water-Energetic-Reactor, VVER) based on $^{103}\text{Ru}/^{106}\text{Ru}$ activity ratios of regular spent nuclear fuels (UO₂ and mixed oxide fuel, MOX) at the end of their fuel lifetimes. The light gray area represents the uncertainty of the ratio.

Laboratory (Italy). The experiment is set up around the liquid scintillator “Borexino” (Italian diminutive of BOREX, Boron Solar Neutrino Experiment). In an experiment called Short Baseline Neutrino Oscillations with Borexino (SOX-Borexino), the existence of a hypothetical fourth (sterile) should be tested by positioning a powerful, yet compact ^{144}Ce - ^{144}Pr source in the vicinity of the Borexino detector to induce nuclear reactions of the inverse β -decay type (36). According to Vivier et al. (37), the Mayak reprocessing facility has been identified as the only potential supplier to have the chemical capabilities to produce a ^{144}Ce antineutrino source with sufficient activity (approximately 3.5 to 5 PBq ^{144}Ce) and purity (38, 39). In this facility, sources are produced using fission product solutions from spent fuel reprocessing. Reprocessing of spent nuclear fuel from VVER-440 reactors for the purpose of production of petabecquerel sources of ^{144}Ce for sterile neutrino experiments is discussed in Gerasimov et al. (40). The authors suggest reprocessing spent fuel with a cooling period of 3 y after irradiation. Apparently, “fresher” fuel with less cooling is not being considered, as the extremely high activity levels in such fuel may cause problems in the reprocessing and handling. For example, western reprocessing facilities, such as La Hague (France), do not consider reprocessing of spent fuel until at least 4 (or even 10) y after irradiation (34, 41). However, shorter irradiation duration of the nuclear fuel (1 or 2 y instead of 3 y) is being discussed as a potential way to yield higher specific activities of ^{144}Ce in the entire (mainly stable) fission-derived cerium fraction. Indeed, the onset of stable Ce in the fuel becomes a crucial factor for the production of such source. While the activity of 3.5 to 5 PBq ^{144}Ce corresponds to 30 to 43 g of this radionuclide only, this amount makes up less than 1% of the total mass of Ce that is being isolated from the spent fuel (40). The SOX experiment required a source of ideally at least 3.7 PBq ^{144}Ce with a total mass of 2.5 kg of ultrapure Ce (including both stable and radioactive nuclides) (42), which is challenging to produce. Previously, other radionuclides (including ^{106}Ru) were considered for the SOX-Borexino experiment (36), but ^{144}Ce was found to be easier to extract from spent nuclear fuel. Spent nuclear fuel reprocessing is usually based on the PUREX method (Plutonium Uranium Recovery by Extraction), in which the UO₂ fuel is chopped into smaller fragments and dissolved in 7 to 7.5 M HNO₃. Most fission products (including Ce) are dissolved in the aqueous raffinate; however, a part of the Ru is oxidized to highly volatile RuO_4 and found in the off-gas, where it has to be captured and treated. Fuel components (U and Pu) are recovered by extraction into a kerosene/tri-*n*-butyl phosphate phase

*O. Saunier, A. Mathieu, D. Didier, O. Masson, J. Dumont le Brazidec, Atmospheric modeling and source reconstruction of radioactive ruthenium from an undeclared major nuclear release. *Proc. Natl. Acad. Sci. U.S.A.*, in review.

before they are further processed and recycled. For the isolation of the cerium fraction in the aqueous phase, complexing displacement chromatography techniques were identified, which would yield a cerium fraction with sufficient purity, which would then be converted to CeO_2 and sintered (43). The final product should have fit into a capsule of <15 cm diameter, compact enough to be considered as a point-like source (43).

A critical factor of the attempted ^{144}Ce source is its required extraordinarily high specific activity. While the specific activity of ^{144}Ce per gram of fission-derived Ce can be increased by 28%, when the VVER-440 fuel is irradiated for 2 y instead of 3 y (and by 76% when irradiated for 1 instead of 3 y) (40), the reduction of the cooling time from 3 to 2 y causes a gain in specific activity by more than 140% (based on data from ref. 34). In fact, the aspired specific activity of at least 3.7 PBq of ^{144}Ce in 2.5 kg of ultrapure Ce can hardly be achieved only by reducing the irradiation duration of the fuel (in theory, this goal is only barely reachable when using 1-y-irradiated fuel), but can easily be achieved by reducing the cooling period from 3 y down to ~2 y (even for 5 PBq sources and even from regular spent fuel at the end of its fuel lifetime). The reduction of the cooling period of the spent fuel may have been regarded as the only feasible way to yield a sufficient activity of ^{144}Ce in a yet reasonably small-sized source volume that was needed for the SOX experiments. If the ^{106}Ru release was indeed linked to the production of the SOX source, these considerations concerning the specific activity of the ^{144}Ce source would explain the young age of the Ru fission products at the time of the release. According to the source term estimates, the estimated release of 250 TBq of ^{106}Ru would correspond to an accidental loss of about 7 to 10% of the ^{106}Ru contained in the amount of VVER-440 fuel (700 to 1,000 kg, corresponding to 2 fuel assemblies) that would be necessary for the production of a 3.5 to 5 PBq ^{144}Ce source (assuming regular spent VVER-440 fuel at the end of its lifetime, based on data from ref. 34 and 2 y of cooling). In any case, the unusually young age of the $^{103,106}\text{Ru}$ of about 2 y after the end of irradiation in a power reactor would match the hypothesis of a release of ^{106}Ru during the production of the ^{144}Ce SOX source. Regarding this production, the fact that the order of the ^{144}Ce source was canceled by the Mayak facility shortly after the $^{103,106}\text{Ru}$ release attracted the attention of the nuclear community and initiated speculations whether both events (the release of $^{103,106}\text{Ru}$ and the cancellation of the ^{144}Ce order) may be linked (35). No finding of our research refuted or disproved the hypothesis of a linkage between the radioruthenium detection episode and the production of a ^{144}Ce source.

If the radioruthenium came from the production of the ^{144}Ce source, and if spent fuel has been reprocessed before reaching the end of its fuel lifetime in reactor, the above age estimate would represent a slight underestimate, as the “youth-indicator” ^{103}Ru would be present in “fresher” fuel in proportionally higher concentrations compared with ^{106}Ru . The nonetheless young age of the $^{103,106}\text{Ru}$ also speaks against the “satellite hypothesis” as such young radionuclide batteries would not typically be available, because of a lack of facilities that can routinely handle such young fuel (possibly with the exception of the Mayak industrial complex). **Particle size and solubility.** The released $^{103,106}\text{Ru}$ was found to be evenly distributed on air filters and no larger $^{103,106}\text{Ru}$ -rich particles were found in autoradiographic imaging (*SI Appendix, Fig. S4*) and scanning-electron microscopic investigations (*SI Appendix, Fig. S5*). Therefore, it may be concluded that the radioruthenium was released in a gaseous or extremely finely dispersed particulate form (particle sizes <1 μm). At room temperature, RuO_4 is an easily melting solid (melting point 25 °C), but due its high vapor pressure and low boiling point (40 °C), it is notorious for being a highly reactive and volatile substance (44), even from solutions (45). It is a highly unstable compound and decomposes to chemically inert RuO_2 (and O_2) in an explosive reaction when heated above 100 °C. The explosive properties may also be

accident-relevant for the present release. Given its high reactivity, a release of highly oxidizing, gaseous RuO_4 would be followed by subsequent reduction or decomposition to RuO_2 on [organic or inorganic (46)] atmospheric dust particles and trapping on their surface. The lack of any traces of concomitant radionuclides in the Ro5 aerosol filters suggests a high degree of fractionation of the $^{103,106}\text{Ru}$ from other radionuclides, which would be achievable by a separation of highly volatile RuO_4 from other, less volatile radionuclides via the gas phase. Knowledge on the ^{106}Ru fractionation (gas/particle) would also have direct consequences on deposition in the environment close to the emission point. This is especially the case for the particulate fraction in case of rain or for species that remain in gaseous form. Various investigations at participating research institutions suggest a radiopurity of the $^{103,106}\text{Ru}$ of up to a factor of 10^4 over concomitant radionuclides. Thus, we conclude that the release most likely involved RuO_4 , which either was released directly or possibly underwent chemical processing for its stabilization (e.g., trapping in NaOH or HCl) before its release into the environment. The fact that no apparent stable element anomalies were found in addition to the release supports the assumption that the release was limited to the isotopes of Ru (16).

In chemical investigations, several research laboratories have found that at least one of the ^{106}Ru species on the filters was highly soluble (about 50% within 10 min) when the filter was immersed in pH neutral water. The insoluble fraction remained insoluble even if the immersion duration was extended to 24 h. The solubility was marginally greater in when a filter fragment was immersed into 1 M HCl solution (24 h), namely to about 60%. The filtrate was filtered through syringe filters with pore sizes of 470 nm and 20 nm, respectively, which reduced the activity in the aqueous phase only marginally, thus excluding the breakthrough of suspended ^{106}Ru -containing particles in the water, which might have pretended dissolution. Assuming that the ^{106}Ru was trapped on air filters in the form of RuO_2 (reduction product of RuO_4), this high degree of solubility comes unexpectedly, at least when the macroscopic chemical behavior of RuO_2 is extrapolated to the sub-macroscopic scale of ultratrace levels of ^{106}Ru . Hence, the results may suggest that at least 2 species are present on the filter materials and that half of the total amount of ^{106}Ru is present in highly water-soluble form. Solubility experiments in serum ultrafiltrate fluid (SUF), however, reveal very high and rapid solubility (>90%) of the ^{106}Ru on air filters, which were brought in contact with SUF to simulate dissolution in the lungs. Two different kinetics were found

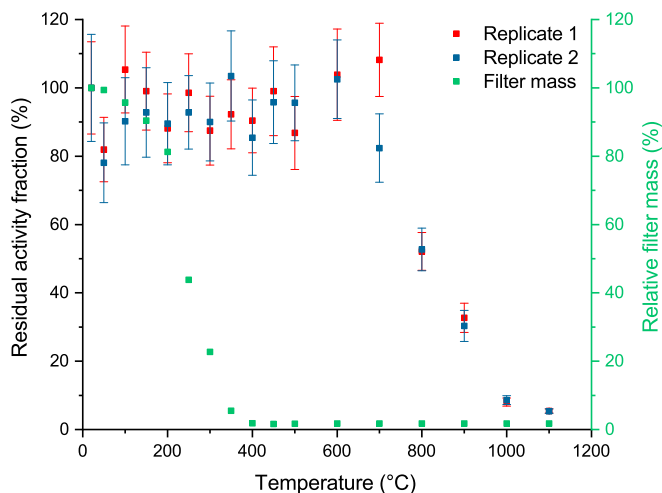


Fig. 7. Volatility tests of ^{106}Ru trapped on an air filter from Vienna (2 replicates with 30 Bq ^{106}Ru pieces). The loss of mass is shown in green for comparison.

to underlie the dissolution (*SI Appendix, Fig. S2*). The latter results may indicate that the Ru is associated with 2 different types of aerosol particles or particle surfaces, 1 of which may be binding ^{106}Ru more strongly than the other. In contrast to H_2O , SUF contains ligands that may out-compete surface binding sites for Ru.

Volatility. In heating experiments, the volatility of the Ru species from the filter was investigated. We found that the ^{106}Ru activities remained quite constant (with some fluctuations due to the thermal deformation of the filter fragments causing geometrical issues during the γ -measurement) (*SI Appendix, Fig. S3*) for the temperature range between room temperature and 600 °C. From 700 °C to 1,000 °C, the activity levels in the filter fragments' ashes decrease rapidly, suggesting almost complete volatilization of the Ru species in this temperature range (Fig. 7). In macroscopic amounts, highly refractory RuO_2 does not exhibit comparable volatilization in this temperature range (47). In combination with the solubility tests, this result suggests that the released Ru species has not been (or not exclusively) RuO_4 , because one would expect low volatility and solubility from its reaction product RuO_2 . Instead, a mixture of several chemical species of ruthenium may have been released.

Summary

Almost exactly 60 y after the Kyshtym and Windscale nuclear accidents in 1957, both substantial sources of ^{106}Ru in the environment (23), a significant release of ^{106}Ru occurred in the southern Eurasian border region in September 2017. It led to detectable yet innocuous levels of this airborne fission product in the rest of Europe. Based on times series of detections at various locations in Central Europe, the event was characterized as a short release. The plume duration lasted about 1 to 3 d on average, depending on the location, with the exception of a few areas (e.g., in Italy where the labeled air mass, once arrived, stayed longer than elsewhere). The release was too substantial to be associated with the incineration of a medical radionuclide source. A satellite crash can also be excluded as the source of the ^{106}Ru . It is much more likely that the ^{106}Ru escaped during reprocessing of spent nuclear fuel, possibly in the course of the miscarried production of a highly radioactive ^{144}Ce source for research applications in the Gran Sasso Laboratory in Italy. This study confirms high radiopurity of the radioruthenium, an age of about 2 y or less, as well as both relatively high solubility and volatility of the Ru species on the air filters. According to detection time series, a back-trajectory analysis, and chemical considerations, the Mayak nuclear complex in southern Urals should be considered as a likely candidate for the release.

Materials and Methods

Aerosol sampling is performed on a routine basis using medium- to high-flow rate pumps (60 to more than 1,000 $\text{m}^3\cdot\text{h}^{-1}$). Airborne radionuclide monitoring on a nation-wide scale is usually based on weekly sampling, but the frequency may be increased by certain stations in unusual events, like the present one. Depending on the network and organization, different kinds of filter media are being used in Europe (glass fibers, glass plus cellulose, polyvinyl chloride, or polypropylene), all having a high (>90%) collection efficiency. After sampling, filters are typically pressed into pellets of various diameters depending both on filter and detector sizes.

^{106}Ru is a pure β -emitter and thus not directly measurable by γ -ray spectrometry. However, its detection can be achieved via decay of its daughter rhodium-106 (^{106}Rh), which reaches secular equilibrium within a few minutes with its parent due to its short half-life ($T_{1/2} = 30.1$ s) and which is detectable by de-excitation γ -photons of the excited state of its decay product: palladium-106 (^{106}Pd , stable). The radionuclide is first identified by its 621.9-keV photopeak (emission intensity 9.87%) in a γ -ray spectrum and, once identified, the presence of ^{106}Ru can be confirmed by characteristic peaks at 1050.4 keV and 616.2 keV. All measurements were thus obtained by γ -ray spectrometry using coaxial or well-type, high-purity germanium detectors. Proper quantification should comply with recommendations in the Joint Research Centre technical report provided by the European Commission (48) regarding interferences and coincidence summing corrections. Indeed, these corrections can be significant, reaching up to deviations of up to 25%, depending on the detector and the counting geometry. For maximum activity concentrations, uncertainties were typically ranging from 5 to ~30% depending on detector and sample features. An example of a γ -ray spectrum exhibiting detectable activities of both ^{103}Ru and ^{106}Ru is shown in *SI Appendix, Fig. S6*. It exemplifies that, thanks to excellent energy resolution in modern γ -ray detectors, the unique γ -ray peaks of ^{106}Ru (^{106}Ru) at 621.9 keV and 616.2 keV are not disturbed by the presence of natural ^{214}Bi (609.3 keV).

Data Materials and Availability. We thank the CTBTO for providing analysis results in the framework of a Virtual Data Exploration Centre agreement (<https://www.ctbto.org/specials/vdec>). IMS raw data collected by the CTBTO are confidential and may not be shared.

ACKNOWLEDGMENTS. We thank all data providers, among them the Environmental Protection Agency (Lithuania) and the Ignalina Nuclear Power Plant Laboratory (Lithuania); the National Oceanic and Atmospheric Administration Air Resources Laboratory, for the provision of the HYSPLIT model for back-trajectory calculations; and the Comprehensive Nuclear-Test-Ban Treaty Organization for providing analysis results in the framework of a virtual Data Exploration Centre agreement (<https://www.ctbto.org/specials/vdec>). We acknowledge financial support by the VolkswagenStiftung (Az. 94403). A.W. acknowledges funding by the Deutschen Bundesstiftung Umwelt. The views expressed in this study are those of the authors and do not necessarily reflect the views of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization.

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