Subscriber access provided by Helmholtz Zentrum Muenchen - Zentralbibliothek

Environmental Modeling

Potential source apportionment and meteorological conditions involved in airborne ^{131}I detections in January/February 2017 in Europe

Olivier Masson, Georg Steinhauser, Herbert Wershofen, Jerzy Wojciech Mietelski, Helmut W. Fischer, Laurent Pourcelot, Olivier Saunier, Jacqueline Bieringer, Thomas Steinkopff, Miroslav Hýža, Bredo Møller, Theodore W Bowyer, Ekaterinini Dalaka, Axel Dalheimer, Anne De Vismes-ott, Konstantinos Eleftheriadis, Maurizio Forte, Catalina Gasco Leonarte, Krzysztof Gorzkiewicz, Zsolt Homoki, Krzysztof Isajenko, Tero Karhunen, Christian Katzlberger, Renata Kierepko, Júlia Kövendiné Kónyi, Helena Malá, Jelena Nikolic, Pavel P. Povinec, Milica Rajacic, Wolfgang Ringer, Petr Rulik, Rosella Rusconi, Géza Sáfrány, Ivan Sykora, Dragana Todorovic, Jochen Tschiersch, Kurt Ungar, and Benjamin Zorko

Environ. Sci. Technol., **Just Accepted Manuscript** • DOI: 10.1021/acs.est.8b01810 • Publication Date (Web): 06 Jul 2018

Downloaded from http://pubs.acs.org on July 16, 2018

Just Accepted

“Just Accepted” manuscripts have been peer-reviewed and accepted for publication. They are posted online prior to technical editing, formatting for publication and author proofing. The American Chemical Society provides “Just Accepted” as a service to the research community to expedite the dissemination of scientific material as soon as possible after acceptance. “Just Accepted” manuscripts appear in full in PDF format accompanied by an HTML abstract. “Just Accepted” manuscripts have been fully peer reviewed, but should not be considered the official version of record. They are citable by the Digital Object Identifier (DOI®). “Just Accepted” is an optional service offered to authors. Therefore, the “Just Accepted” Web site may not include all articles that will be published in the journal. After a manuscript is technically edited and formatted, it will be removed from the “Just Accepted” Web site and published as an ASAP article. Note that technical editing may introduce minor changes to the manuscript text and/or graphics which could affect content, and all legal disclaimers and ethical guidelines that apply to the journal pertain. ACS cannot be held responsible for errors or consequences arising from the use of information contained in these “Just Accepted” manuscripts.

is published by the American Chemical Society. 1155 Sixteenth Street N.W., Washington, DC 20036

Published by American Chemical Society. Copyright © American Chemical Society. However, no copyright claim is made to original U.S. Government works, or works produced by employees of any Commonwealth realm Crown government in the course of their duties.

	Povinec, Pavel P.; Comenius University, Slovakia Rajacic, Milica; Institut za nuklearne nauke Vinca Laboratorija za zastitu od zracenja i zastitu zivotne sredine Ringer, Wolfgang; AGES - Österreichische Agentur für Gesundheit und Ernährungssicherheit GmbH, Rulik, Petr; Statni Ustav Radiacni Ochrany Rusconi, Rosella ; Azienda Regionale per la protezione dell'ambiente Lombardia Sáfrány, Géza ; Orszagos Sugarbiologiai es Sugaregeszsegugyi Kutato Igazgatosag Sykora, Ivan; Univerzita Komenskeho v Bratislave Todorovic, Dragana; Institut za nuklearne nauke Vinca Laboratorija za zastitu od zracenja i zastitu zivotne sredine Tschiersch, Jochen; Helmholtz Zentrum Munchen Deutsches Forschungszentrum fur Umwelt und Gesundheit Ungar, Kurt; Health Canada Zorko, Benjamin; Jožef Stefan Institute,

Potential source apportionment and meteorological conditions involved in airborne ^{131}I detections in January/February 2017 in Europe

Authors

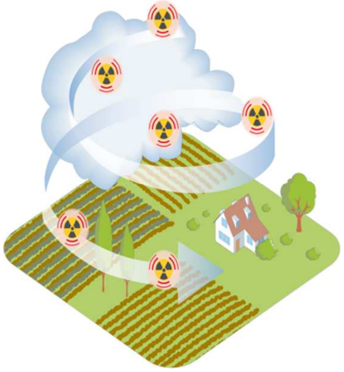
O. Masson*[1], G. Steinhauser[2], H. Wershofen[3], J.W. Mietelski[4], H.W. Fischer[5], L. Pourcelot[1], O. Saunier[1], J. Bieringer[6], T. Steinkopff[7], M. Hýža[8], B. Møller[9], T.W. Bowyer[10], E. Dalaka[11], A. Dalheimer[7], A. de Vismes-Ott[1], K. Eleftheriadis[11], M. Forte[12], C. Gasco Leonarte[13], K. Gorzkiewicz[4], Z. Homoki[14], K. Isajenko[15], T. Karhunen[16], C. Katzlberger[17], R. Kierepko[4], J. Kövendiné Kónyi[14], H. Malá[8], J. Nikolic[18], P.P. Povinec[19], M. Rajacic[18], W. Ringer[20], P. Rulík[8], R. Rusconi[12], G. Sáfrány[14], I. Sykora[19], D. Todorović[18], J. Tschiersch[21], K. Ungar[22], B. Zorko[23]

* olivier.masson@irsn.fr

Affiliations

1 Institut de Radioprotection et de Sûreté Nucléaire, (IRSN), Fontenay-aux-Roses, 92262, France
2 Leibniz Universität Hannover, Institute of Radioecology and Radiation Protection, Hannover, 30419, Germany
3 Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, 38116, Germany
4 The Henryk Nievodniczanski Institute of Nuclear Physics, Polish Academy of Sciences (IFJ), Kraków, 31-342, Poland
5 University of Bremen, Institute of Environmental Physics, Bremen, 28359, Germany
6 Bundesamt für Strahlenschutz (BfS), Freiburg, 79098, Germany
7 Deutscher Wetterdienst (DWD), Offenbach, 63067, Germany
8 National Radiation Protection Institute (NRPI), Prague, 140 00, Czech Republic
9 Norwegian Radiation Protection Authority (NRPA), Svanvik, NO-9925, Norway
10 Pacific Northwest National Laboratory (PNNL), P.O. Box 999, Richland, WA 99352, USA
11 Institute of Nuclear and Radiological Sciences & Technology, Energy & Safety, NCSR "Demokritos", Environmental Radioactivity Laboratory, Ag. Paraskevi, Attiki, 15310, Greece
12 Agenzia Regionale per la Protezione dell'Ambiente (ARPA Lombardia), Milan, 20129, Italy
13 Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT), Madrid, 28040, Spain
14 "Frédéric Joliot-Curie" National Research Institute for Radiobiology and Radiohygiene, (OSSKI), POB 101, Budapest, H-1775, Hungary
15 Central Laboratory for Radiological Protection (CLOR), Warsaw, PL 03-134, Poland
16 Radiation and Nuclear Safety Authority (STUK), P.O. Box 14, Helsinki, 00811, Finland
17 Radiation Protection and Radiochemistry, Austrian Agency for Health and Food Safety (AGES), Wien, 1220, Austria
18 Vinča Institute of Nuclear Sciences, Belgrade, 11001, Serbia
19 Comenius University, Department of Nuclear Physics and Biophysics, Bratislava, 84248, Slovakia
20 Radioecology and Radon, Austrian Agency for Health and Food Safety (AGES), Linz, 4020, Austria
21 Helmholtz Zentrum München, German Research Center for Environmental Health (HMGU), Neuherberg, 85764, Germany
22 Health Canada (HC-SC), Radiation Protection Bureau, Ottawa, A.L. 6302A, Ontario, K1A 1C1, Canada
23 Jozef Stefan Institute (IJS), Ljubljana, 1000, Slovenia

ABSTRACT: Traces of particulate radioactive iodine (^{131}I) were detected in the European atmosphere in January/February 2017. Concentrations of this nuclear fission product were very low, ranging 0.1 to 10 μBq m^{-3} except at one location in western Russia where they reached up to several mBq m^{-3}. Detections have been reported continuously over an 8-week period by about thirty monitoring stations. We examine possible emission source apportionments and rank them considering their expected contribution in terms of orders of magnitude from typical routine releases: radiopharmaceutical production units > sewage sludge incinerators > nuclear power plants > spontaneous fission of uranium in soil. Inverse modeling simulations indicate that the widespread detections of ^{131}I resulted from the combination of multiple source releases. Among them, those from radiopharmaceutical production units remain the most likely. One of them is located in Western Russia and its estimated source term complies with authorized limits. Other existing sources related to ^{131}I use (medical purposes or sewage sludge incineration) can explain detections on a rather local scale. As an enhancing factor, the prevailing wintertime meteorological situations marked by strong temperature inversions led to poor dispersion conditions that resulted in higher concentrations exceeding usual detection limits in use within the informal Ring of Five (Ro5) monitoring network.



INTRODUCTION

Radioactive ^{131}I is an important fission product that is produced with high yield by nuclear fission of ^{235}U (2.9%) or ^{239}Pu (3.8%). The half-life of ^{131}I is short enough (8.02 days) to cause high specific activities, but long enough to allow long-distance (> 3,500 km) atmospheric dispersion when released into the atmosphere. Elementary iodine (I_2) is a solid under normal conditions of temperature and pressure and slowly sublimates even at room temperature leading to the formation of gaseous I_2. Due to its volatility and reactivity with many compounds (aerosol, ozone, volatile organic compounds, etc.) iodine in the ambient atmosphere is distributed in variable proportions between gaseous and particulate species. The ratio between the various released chemical forms may change along the route of air masses and from daytime to nighttime as a result of photo-dissociation. Even if the release occurs only in gaseous form, iodine in air can transform to less volatile aerosol-bound species or attach to existing particles.[1, 2] All those properties make ^{131}I one of the most important radionuclides for the monitoring of airborne radioactive releases.

The topic of this study is the early 2017 detection of airborne ^{131}I at trace levels in Europe that made headlines in media and that spread rapidly on social networks[3] after the publication of a single map gathering all detections until the first week of February.[4] Here, we report detailed time trend and geographical evolution of airborne ^{131}I concentrations and evaluate several potential sources based on authorized releases or real releases when available. Several significant emissions of ^{131}I from radiopharmaceutical production units have already been reported in the literature during the last decade leading to a similar widespread detection of ^{131}I at trace levels on the European scale.[5-10] Other local sources such as sewage plant incinerators, nuclear power plants, as well as possible ubiquitous and natural production of ^{131}I from the spontaneous fission of uranium in soils, have also been examined. Finally, we investigated the meteorological conditions as a "detection enhancing factor" using airborne naturally occurring radionuclides (namely ^{7}Be, ^{22}Na, ^{210}Pb, and ^{40}K), an anthropogenic radionuclide (^{137}Cs), and particulate matter with an aerodynamic diameter <10 µm (PM_{10}) values as indicators of atmospheric dispersion by comparing their values during the ^{131}I detection weeks with their usual background levels.

Most reported data come from atmospheric monitoring stations gathered within the "Ring of Five" (Ro5) network which is an informal collaboration platform on laboratory level.[11] According to our experience based on internal Ro5 reports, trace detections of unexpected radionuclides occur on a more frequent basis than what one might anticipate. On average, there is a handful of unexpected or unusual radionuclides detected at trace-level each year on a European-wide scale and a dozen on a local scale. When only one station detects unexpected radionuclides or enhanced activity concentrations at trace levels, it is a general practice within the Ro5, not to spread the information outside the country because it may be representative of only a local activity with an isolated impact. However, in some cases and even for a single detection, it may be worth sharing this information with the Ro5 community, especially when there is no apparent reason for an anticipated abnormality at this location, for example when faraway from any nuclear or industrial facility. This was the case in January 2017, when ^{131}I was first detected in the Norway's northeasternmost corner. Most detections reported by Ro5 members proved to be very close to the minimum detectable activity (MDA), in the µBq m^{-3} range. Given the great public concern regarding releases of anthropogenic radionuclides, the policy of non-publicizing very localized trace concentrations in the air is normally carried out as long as levels are of no health concern. However, on few occasions, radionuclide detections simultaneously occur at several monitoring stations, and even if at innocuous levels, these detections should become public.[1, 12] In the late fall of 2011, the attention of the Ro5 members was triggered by the detection of ^{131}I in the European atmosphere as well as in animal thyroid biomonitors after its release from a radiopharmaceutical facility in Hungary.[13, 14] Other multiple detection events of particulate ^{131}I at trace levels were also reported within the Ro5, especially in Scandinavia, in January/February 2012, March 2015, October 2016, and recently in January until beginning of March 2018.[15]

Materials and Methods

The monitoring stations gathered within the Ro5 network operate according to national regulations and are usually equipped with high-volume air samplers having an air throughput rate of several hundred m^3 h^{-1} and up to 1,500 m^3 h^{-1}. Aerosols are usually sampled on a weekly basis (traditionally from Monday to Monday). Iodine-131 counting is performed by gamma spectrometry, using low-level High-Purity Germanium (HPGe) detectors and identifying ^{131}I by its 364 keV line in the gamma spectrum. Recent spectrometry improvements (new shields, Compton-suppression systems, higher detection efficiency, etc.) made it possible to lower the MDA for particulate ^{131}I down to 0.5 μBq m^{-3}, and even 0.1 μBq m^{-3} when using very high-volume samplers. This concentration corresponds to the staggering amount of 1 atom of ^{131}I per 10 m^3 of air. Such MDA currently in use within the Ro5 network, allow quantifying concentration levels, which had yet remained undetectable only five years ago.

Results and Discussion

Sequence of airborne ^{131}I detections in Europe.

The first ^{131}I detections were reported for Week 2 (January 9-16, 2017) by the Norwegian Radiation Protection Authority at the northernmost European aerosol sampling location in Norway (Svanvik); by the Finnish Radiation and Nuclear Safety Authority in the northern part of Finland (Rovaniemi) and by the Central Laboratory for Radiological Protection on the same week in Poland (Warsaw). Over the entire detection episode (Week 2 to Week 8), 38 particulate ^{131}I detections above MDA were reported from 26 locations in Europe (Fig. 1). One unique detection of gaseous ^{131}I (10.6 ± 3.4 μBq m^{-3}) was reported on Week 4 by the laboratory of the "Centro Regionale Radioprotezione" (Arpa Lombardia) in Milan (Italy) thanks to a high-volume gaseous sampler operating at about 80 m^3 h^{-1}. Indeed, detection limits for gaseous ^{131}I are usually at least 100 times higher than for the aerosol fraction, mainly because most commercially available charcoal cartridges do not allow for such a high throughput rates of air (typically between 3 and 10 m^3 h^{-1}) like for the aerosol samplers. Unfortunately, the aerosol-bound ^{131}I fraction at that location was not detected above a detection limit of 5 µBq m^{-3}, thus preventing the determination of the gas-to-particle ratio. However, inasmuch as a comparison can be made with the situations in Europe after the Chernobyl and Fukushima accidents, we assume a similar gas-to-aerosol ratio between 3 and 5.[1] For the present event, this assumption leads to a particulate iodine concentration of about 2 to 3 µBq m^{-3}, which would be below the detection limits of the measurement laboratory in question.

All in all, weekly ^{131}I aerosol concentrations remained in the µBq m^{-3} range, except in Hungary (Budapest) where the level peaked momentarily at 9.8 µBq m^{-3}, and in western Russia where daily particulate ^{131}I levels rose up to several tens µBq m^{-3} in Dubna and up to 3 orders of magnitude higher (mBq m^{-3} range) in Obninsk (at about 100 km southwest of the Moscow city center), thus indicating a high concentration gradient in this region.[16] The Dubna station belongs to the International Monitoring System (IMS) supporting the Comprehensive nuclear Test Ban Treaty Organization (CTBTO).[17, 18] Due to restrictions implemented by confidentiality agreements, we cannot provide the raw data for the IMS station. The Obninsk station is managed by the "Typhoon" Research and Production Association (Typhoon RPA) in connection with the Russian Federal Service for Hydrometeorology and Environmental Monitoring (Roshydromet).

Apart from these two Russian locations, no significant concentration gradient could be observed over the rest of Europe, which would have helped pointing out the main source. Even when considering that the gaseous fraction was omitted and that its contribution would have led up to a five-time higher concentration, ^{131}I total concentrations would have been so low that they raised no health concern. They corresponded to 1/1,000 or even 1/10,000 of those observed in 2011 when the diluted

Fukushima-labeled air masses were distributed over Europe, which themselves were of no health concern.[1]

Potential sources of airborne iodine

The main feature of this event is the wide spreading of ^{131}I on the European scale. Even if the concentrations were very low, they were all in the same range as if they resulted from the dispersion from a more or less remote source emission or from a combination of several emissions from different locations. Even if it may have been confined within annual authorization limits, it can also be considered that the plume rooted in a significant release of radionuclides. For instance, the European-wide ^{131}I detection event in fall 2011 resulted from a release of 342 GBq from the Institute of Isotopes near Budapest (Hungary) and led to somewhat similar, even if slightly higher, ^{131}I concentrations (0.5 - 68 µBq m^{-3} outside Hungary).[8, 14] Source apportionment and ranking in terms of contribution to the observed values are detailed hereafter.

Figure 1

Hypothesis for the origin of airborne ^{131}I: Research reactors or radiopharmaceutical facilities

In nuclear medicine, most diagnostic procedures rely on ^{99m}Tc whose precursor, ^{99}Mo, is produced using low or highly enriched uranium targets.[19] Iodine-131 can be considered as a by-product of the ^{99}Mo production.[20] Table 1 summarizes the major ^{99}Mo, and thus ^{131}I, suppliers in Europe and Western Russia. All these facilities have yearly iodine release authorizations ranging between one GBq and up to several hundred GBq. To cope with an increasing demand for radiopharmaceuticals (see following hypothesis), there is an increasing effort since several years to increase radiopharmaceutical production capabilities. For instance in 2015, the ^{131}I release (850 GBq) from the L. Ya. Karpov Institute of Physical Chemistry (Obninsk branch) and hereafter named “Karpov Institute”, reached and exceeded its yearly authorization limit (780 GBq) to fulfil the needs and this limit was about 5 times higher than that in 2014.[21] Many other facilities are producing medical radionuclides in Russia[22] but regarding the frequent detections in Obninsk, they are closely related (r = 0.9) to the release of radionuclides from the Karpov Institute in Obninsk.[16, 23]

Table 1. Main ^{99}Mo/^{131}I producers in Europe and Western Russia.

Country (Town)	Company or Institute	Maximum yearly authorized ^{131}I release to the atmosphere (GBq)	Reference
Poland (Otwock-Świerk)	Nuclear Research Radioisotope Centre Polatom	0.1 in 2016	24
Netherlands (Petten)	Mallinckrodt Medical B.V	0.3 in 2017	25
France (Saclay)	UPRA (Cis-BIO international)	0.6 in 2013	26
Belgium (Fleurus)	Institut national des RadioEléments (IRE)	41.8 in 2011	10
Russia (Obninsk)	L. Ya. Karpov Institute of Physical Chemistry (NIFKhI)	780 in 2015	21
Hungary (Budapest)	Institute of Isotopes Ltd (IoI)	1600 in 2011	27

Apart from the main radiopharmaceuticals producers, there is an increasing number of secondary industries that transform concentrated ^{131}I solutions into diluted ones (injectable or for oral administration) as well as into ^{131}I capsules, which may also represent a potential source of ^{131}I emission. Despite significant amounts of ^{131}I released within authorized limits, airborne detections of ^{131}I coming from major radiopharmaceutical production units remain rare and unusual. Only close monitoring stations may report regular detections at trace concentrations that exhibit negligible doses upon inhalation for the local population.[23] Several significant ^{131}I emissions from radiopharmaceutical production units have already been reported in the literature as a result of incidents during the last decade, leading to a similar widespread detection of ^{131}I at trace levels (tenths to tens of µBq m^{-3}) on the European scale.[8, 9, 14, 27] In August 2008, an incident release of ca. 48 GBq of gaseous molecular ^{131}I occurred at the Institute of RadioElements (IRE) in Belgium.[8,9] This amount was released at once and corresponded to the yearly ^{131}I release authorization. The incident was rated 3 on the International Nuclear and radiological Event Scale (INES). Unfortunately, this event is not documented with regard to measurements in the atmosphere. Since this incident, the releases have been reduced by about a factor of 10 (IRE Staff, personal communication). The second event concerns the release of 342 GBq of ^{131}I from the Institute of Isotopes Ltd., Hungary in the time interval between September 8 and

November 16, 2011.[14, 27] This incident release was rated 1 on the INES scale. It led to detections in the range of a few to several tens of µBq m^{-3} in Europe, not only by Ro5 stations but also by aerosol stations of the International Monitoring Systems (IMS). These stations belong to the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) and are instrumental for the monitoring of the planet for clandestine nuclear explosions. During 2008 (with 55 of 80 stations being operational), 6.0 % of all detections within the IMS network were attributed to ^{131}I. For example, the IMS station RN61 in Dubna, Russia, has observed “regular emissions from nearby medical or nuclear facilities” in the past.[6]

From the box-and-whisker plot shown in Fig. 1, it is also noteworthy mentioning that the “highest” particulate ^{131}I concentrations reported in Europe during the studied event came from monitoring stations located in the vicinity of radiopharmaceutical production units. This was the case at several locations: in Warsaw (on Week 2) which is ca. 30 km from the Polatom research Centre; in Budapest (Weeks 4 and 7) which is 9 km away from the Institute of Isotopes and in Obninsk where the Karpov Institute is located, with 2.4 mBq m^{-3} from January, 13 to 16; and 2.1 mBq m^{-3} from February, 3 to 6.[28, 29]

Hypothesis for the origin of airborne ^{131}I: Nuclear medicine hospitals and sewage sludge incineration plants

Some aerosol sampling stations that detected small traces of ^{131}I are located in the vicinity of a nuclear medicine hospital or a sewage sludge plant that collects their waste waters. This was the case in Freiburg (Germany), Krakow (Poland) or in České Budějovice (Czech Republic). Some national monitoring bodies or local institutes in charge of the surveillance of the atmosphere were not surprised to detect minute amounts of ^{131}I on their filters because such detections are known to occur sporadically, especially in winter. Involvement of sewage sludge incinerators on the regular airborne ^{131}I detection has already been pointed out at short distance.[30]

Iodine-131 is a frequently used radionuclide in nuclear medicine with therapeutic (rather than diagnostic) applications. The administered activities vary with the purpose of the treatment. Hyperthyroidism is treated with lower activities (0.2 – 0.5 GBq ^{131}I per treatment) than thyroid cancer (1.8 – 9.2 GBq per treatment).[6] In 1996, some 180,000 patients were treated on a worldwide basis for hyperthyroidism and another 20,000 for thyroid cancer.[6] The projected requirement for ^{131}I in the USA for thyroid cancer treatment in 2009 is 170 TBq of ^{131}I. [6] The widespread use and growing demands for imaging organ function and disease states or treatment are directly attributable to the development and availability of a vast range of specific radiopharmaceuticals.[22, 31] Application rates of ^{131}I in other countries in recent years illustrate the extent of its use: Argentina: 300 TBq y^{-1}; Bangladesh: 5 TBq y^{-1}; Chile: 15,000 patients per year requiring 15 TBq y^{-1}; India: 60 TBq y^{-1}; Thailand: requiring 15 TBq in 2008; Austria: 1 TBq in 2008; Australia: approx. 200 patients per year requiring 4 - 6 GBq per patient. [6] A fraction of the ^{131}I activity administered to the patient is excreted into the sewer and transported to water purification plants.[32, 33] This process takes place even when patients are hospitalized and their excrements (at least urines) are collected for decay, as it is almost unavoidable that the patients still carry a residual activity of multiple MBq at the time of release from the hospital. In France, urines are stored for decay in dedicated tanks but faeces are drained out without significant decay because of the nosocomial hazard that has to be avoided within hospitals. German regulations allow patient release from hospitals below a residual ^{131}I body activity of 250 MBq.[34] Radioactive waste releases from hospitals are subject to authorizations. For instance, in Germany ^{131}I concentrations in airborne releases are limited to 0.5 Bq m^{-3}, and in liquid effluents to 5 Bq L^{-1}. [35] However, except some local or national initiatives such as the one conducted in Germany in the framework of the Precautionary Radiation Protection Act, there is no European Union (EU) regulation with regard to release of radionuclides from sewage sludge incinerators, as they are not considered as radiation controlled areas. Exemption levels for large amounts of solid materials are comparatively high, e.g. 10^4 Bq kg^{-1} of ^{131}I according to the new European radiation protection legislation.[36]

Only a fraction of the ^{131}I reaching the sewage plant is retained in sewage sludge, since radioiodine remains mainly in aqueous solution.[37] Nevertheless, routine monitoring results for Germany reveal median ^{131}I concentration in digested sludge $C_{\text{I-131(sludge)}}$ of 30 Bq kg^{-1} (dry mass).[38, 39] The solid phase from the purification of waste water is known for its high content of several undesired ingredients such as heavy metals, pesticides, dioxins, viruses and bacteria,[39] but the content of traces of radioactive materials from medical applications is rarely considered with regard to potential release, and except for dose assessment for workers exposed inside the sewage treatment plant. The presence of poisons reduces the applicability of this material as fertilizers, so in recent years the combustion of sewage sludge has been promoted. The EU recommends the use of sewage sludge as an alternative fuel for energy production by combustion. This energy can be used on-site to optimize the energetic yield of the incinerator, for sludge drying or for heating of the premises. It also avoids or minimizes the costly transportation and storage outside the plant. Total annual production of dry sewage sludge in the 27 countries of the EU currently reaches almost 10^7 t, of which a variable percentage is incinerated. [40] For example, in Poland, only 3 % of the 160,000 t of dry sludge that could have been burnt were actually incinerated in 2009,[41] but this rapidly increased to 8.7 % in 2011, with aims to increase the proportion to 30 % by 2020.[42] In Germany, the percentage of sludge being annually incinerated has increased by 20 % from 1999 to 2001 corresponding to ~ 0.46×10^6 t, to about 60 % of the totally produced sludge amount in 2013 and 2014, and corresponding a sewage sludge sum up $m_{(\text{sludge per year})}$ of 10^6 t (dry mass).[42,43] High combustion percentages of sewage sludge have been reported also for Belgium, Austria, France, the highest being for Switzerland (97 %).[42]

In all large sewage plants, the sludge designated for incineration is burnt almost immediately to avoid the necessity of interim storage space (Bremen WWTP administration, personal communication). Therefore, no significant decay of ^{131}I needs to be considered and contributes to the release of ^{131}I into the atmosphere despite the filtration system efficiency. In order to reduce odor emissions, sewage sludge mono-incineration facilities operate at temperatures between 850 °C and 950 °C.[45] This temperature range is far above the boiling points of methyl iodine (42.4 °C) and molecular iodine (184.4 °C) leading to the two main gaseous species considered with respect to ^{131}I and its radiological impact. Modern incineration plants filter out dust particles from the off-gases and apply scrubbing to the vapors. IRSN has performed measurements (not published yet) of ^{131}I concentration both in wastewaters entering a sewage treatment plant and released by its sewage sludge incinerator. The average concentration in the liquid waste (0.4 Bq L^{-1}) at an average flow rate (2,500 m^3 h^{-1}) results in 10^6 Bq h^{-1}. Measurements made on samples taken in the flue gas of the sewage sludge incinerator after the fumes had passed through an electro-filter and a charcoal trap, lead to a release of 7×10^3 Bq h^{-1} corresponding to ~ 1% of the amount of ^{131}I entering the plant. For the need of establishing orders of magnitude an emission factor (e_f) ranging 1 to 10 % and corresponding to the capture efficiency of gas and fly ash filtering, can be considered. The total ^{131}I release to the lower atmosphere by sewage sludge incineration for recent years can be calculated to be:

$$E_{\text{I-131(year)}} = C_{\text{I-131(sludge)}} \cdot m_{\text{(sludge per year)}} \cdot e_f \qquad (1)$$

With the previously mentioned values for Germany ($C_{\text{I-131(sludge)}}$ = 30 Bq kg^{-1}, $m_{\text{(sludge per year)}} = 10^6$ tonnes and e_f = 1 to 10%) this results in a yearly release of 0.3 to 3 GBq of ^{131}I to the lower atmosphere, or 0.8 to 8 MBq on a daily basis.

The residence time of the released radioiodine will be lower than its radiological lifetime of τ = 11.57 d, due to deposition processes. As an estimate, we use the residence time, which can be extracted from the records of Fukushima-originating ^{131}I concentration in the ground level air, obtained in Germany in 2011. Records from the German Bundesamt für Strahlenschutz (BfS)[45] show an almost exponential decrease over several weeks, from which an effective residence time τ_{eff} between 4 and 6 days can be extracted. A slightly lower value (3 to 5 d) was found over the North Pacific Ocean for the iodine emissions from the Chernobyl accident.[2] Taken an average value of 5 days, the total ^{131}I activity in the atmosphere over Germany is then given by

$$A_{\text{tot, I-131}} = E_{\text{I-131 (day)}} \cdot \tau_{\text{eff}}. \qquad (2)$$

Using $\tau_{\text{eff}} = 5$ d, we get $A_{\text{tot, I-131}}$ = 4 to 40 MBq.

Assuming a height of the atmospheric mixing layer of h_m = 1,000 m, the average ^{131}I activity concentration in air over Germany can be obtained by

$$a_{\text{I-131}} = A_{\text{tot, I-131}} / (F_{\text{Germany}} \cdot h_m) \qquad (3)$$

which, with $F_{\text{Germany}} = 3.6 \times 10^{11}$ m^2, results in $a_{\text{I-131}}$ = 0.01 to 0.1 µBq m^{-3}.

Hypothesis for the origin of airborne ^{131}I: Nuclear Power Plants

Nuclear power plants have ^{131}I authorization for their release to the atmosphere. Filter and gas trap efficiency are regularly tested to verify that the equipment complies with the requirement of a minimum scrubbing efficiency (upstream [^{131}I] / downstream [^{131}I] > 1000) for the gaseous fraction. The French situation (58 PWR reactors with individual electrical power ranging 900 to 1,450 MWe) corresponds to the highest possible expected airborne concentrations and has thus been taken as a maximum source contribution. The sum of the reported ^{131}I releases to the atmosphere from all French

NPPs during 2013 leads to 0.3 GBq y^{-1}. Note that airborne monitoring performed in the environment close to these NPPs in normal operation has never revealed ^{131}I concentration above detection limits and such releases are supposed to occur mainly during the filter efficiency tests performed with CH_3-^{131}I, but have been considered here like a chronic release. Taking a one-day release corresponds to 0.8 MBq and assuming an effective ^{131}I residence time τ_{eff} of 5 days as for equation (2) leads to $A_{tot,\,I\text{-}131}$ = 4 MBq. Assuming a height of the mixing layer of h_m = 1000 m, the average total ^{131}I activity concentration in air over France can be obtained by

$$a_{I\text{-}131} = A_{tot,\,I\text{-}131} / (F_{France} \cdot h_m) \qquad (4)$$

which, with F_{France} the metropolitan area = 5.5×10^{11} m^2 results in $a_{I\text{-}131}$ = 0.007 µBq m^{-3}. Proper comparison with the observations performed in January/February 2017 requires considering a particle-to-gas ^{131}I ratio of about 30%, based on previous works[1] and leads to 0.0022 µBq m^{-3} for the expected ^{131}I particulate fraction, which is out of reach for the most sensitive gamma-ray detection systems.

Hypothesis for the origin of airborne ^{131}I: Spontaneous fission-derived ^{131}I from natural ^{238}U

There is plenty of literature dedicated to emanation of geogas from soil,[47] including radon (^{222}Rn).[48] Trace amounts of volatile fission products can also be produced by spontaneous fission of naturally occurring heavy nuclides. This has already been demonstrated for ^{129}I, however, it has never been evaluated for ^{131}I, so far. Of all relevant nuclides, ^{238}U has the highest spontaneous fission rate with 5.5 $\times 10^{-5}$ % of the entity of disintegrations.[49] The average concentration of U in the upper continental crust is 2.8 mg kg^{-1}, the vast majority of which is ^{238}U (99.27 %).[50]

The *in situ* production of some fission products by spontaneous fission of ^{238}U within the rock has been estimated and compared with ^{129}I measured in the Stripa granite, located about 170 km WNW of Stockholm, and characterized by an unusual high uranium content (44 ppm) of more than ten times the average value for granite.[50, 51] However, there is no available estimation reporting the subsurface production of ^{131}I and its potential emission to the atmosphere. Assumptions about an underground source of ^{131}I are deduced by analogy with ^{129}I or ^{222}Rn produced by fission and decay of ^{238}U, respectively before their exhalation (Table 2).

In the following, the fission product release estimation is carried out for rock representative of the earth's upper crust, characterized by a rather low average ^{238}U activity of 30 Bq kg^{-1}. For such concentrations, the induced fission of ^{235}U has been neglected. The *in situ* production of ^{131}I by the spontaneous fission of ^{238}U is estimated using equation (5) initially derived for ^{129}I from [46] which gives the equilibrium number of ^{131}I atoms in 1 cm^3 of rock, (^{131}N$_e$):

$$^{131}\mathrm{N_e} = \frac{({}^{238}N\ {}^{238}\lambda_{sf}\ {}^{131}Y_{sf})}{{}^{131}\lambda} \qquad (5)$$

where ^{238}N is the number of ^{238}U atoms per cm^3, $^{238}\lambda_{sf}$ and $^{131}\lambda$ are the spontaneous decay constants of ^{238}U and ^{131}I, respectively and ^{131}Y$_{sf}$ is the spontaneous fission yield of ^{131}I.

Using the same equation for ^{129}I leads to a concentration of activity in the rock of 1.29×10^{-5} Bq m^{-3} which is lower than the estimation for Stripa granite (2.34×10^{-4} Bq m^{-3}) (Table 2). [52] This difference results from the higher ^{238}U activity in the granite (550 Bq kg^{-1}) compared with the average upper continental crust value of 30 Bq kg^{-1}. [51] Furthermore, the comparison of the activity of both *in situ* produced iodine isotopes shows that the production of ^{131}I is larger than that of ^{129}I by about a factor of 20, due to a higher production rate for ^{131}I. Thereafter, the loss of iodine from minerals is mainly due to the recoil of radionuclides. The high activity level recorded in the Stripa granite water is accounted by the diffusion of iodine in the rock porosity and the dissolution in the underground water.[52] Even so, these levels are less than 1 % of the estimated concentrations due to *in situ* production and total diffusive loss from the granite to the water.[51] Similar reasoning can be applied for other primordial radionuclides such as ^{232}Th (spontaneous fission) and ^{235}U (induced fission). At a first estimate, the consideration of thorium leads to the same order of magnitude as for uranium.

The transfer of fissiogenic iodine from subsoil to the atmosphere can be simplified as follows: once produced in the rock, volatile iodine diffuses to the soil surface. During the diffusion stage, volatile iodine will encounter soil organic matter that will considerably reduce the emission to the atmosphere. Iodine sorption on natural organic matter remains complex.[53] However, in case of anoxic conditions (e.g. peat bog or water saturated area), poorly sorbed iodine species and volatile iodine species driven by microbial processes can be emitted to the atmosphere.[54] In the case of ^{131}I, its rather short half-life will also compete with the diffusion duration and will limit the activity that will effectively be emitted to the atmosphere.

The ^{131}I activity measured in the atmosphere soon after underground fission nuclear tests shows that less than 1 % of ^{131}I reaches the surface.[55] Further isotopic data from this study shows that there is no significant fractionation between the xenon isotopes and the precursors, namely iodine isotopes, suggesting that iodine could behave like a noble gas during diffusion to the surface. Following this assumption, we use the ratio between activity of ^{222}Rn in the rock (7.8×10^4 Bq m^{-3}) and the radon activity typically recorded in the troposphere (10-30 Bq m^{-3}) to derive the maximum activity of ^{131}I in the atmosphere, ranging between 3.6×10^{-8} and 1.1×10^{-7} Bq m^{-3}, respectively (Table 2). However, considering the strong sorption of iodine into soils and interaction with organic matter, about 1 % only of radioiodine can reach the surface.[56] This leads finally to ^{131}I activity in the troposphere of ca.10^{-9} Bq m^{-3}. This is far below the MDA of the highest performing monitoring stations.

Table 2: Calculation of the in situ production of ^{131}I and ^{129}I by spontaneous fission of ^{238}U in the rock and estimation of the activity in the troposphere. In situ production of ^{222}Rn from ^{238}U decay is also calculated, assuming secular equilibrium among U-daughters.

		^{222}Rn	^{131}I	^{129}I
calculation parameters	Process of *in situ* production	decay of ^{238}U	spontaneous fission of ^{238}U	spontaneous fission of ^{238}U
	Production half-life (y)	4.47×10^{9}	8.20×10^{15}*	8.20×10^{15}*
	Production yield (%)	100	0.65**	0.03**
	Decay period of produced radionuclide (y)	1.04×10^{-2}	2.20×10^{-2}	1.57×10^{7}
calculation results	*In situ* activity (Bq m^{-3})	7.8×10^{4}	2.8×10^{-4}	1.3×10^{-5}
	Tropospheric concentration (Bq m^{-3})	10-30	1.1×10^{-9}	5.4×10^{-11}

* ref.[57]
** refs.[58] and [59]

Source apportionment

The above estimates for the various ^{131}I source hypotheses are ranked in Fig. 2. It is clear that, given the current MDA, any contributions of ^{131}I either from spontaneous fission of uranium or thorium in soil or from nuclear power plants remain undetectable by the high-volume sampler and low-level detection equipment of the Ro5 members. The situation is similar for the release source from sewage sludge incinerators on a global scale but this explanation for detection on a local scale cannot be ruled out. The most likely release source may concern radiopharmaceutical production units or handling sites. The release of 342 GBq of ^{131}I that took place in Hungary in fall 2011 resulted in a detectable activity range of particulate ^{131}I about 5 to 10 times higher than the one observed in January/February 2017. Assuming the same dispersion coefficient for both events leads to a rough estimate of the source term that may have been released in January 2017 of about one tenth, thus about 35 to 70 GBq (Fig. 2). Note that this amount typically matches a monthly average ^{131}I authorized release of 65 GBq derived from the yearly authorization (780 GBq y^{-1} / 12 months) from the Karpov Institute alone.

Figure 2

Inverse modeling and attempted localization of the origin of the release

Considering the likelihood of radiopharmaceutical unit involvement, inverse modeling techniques have been applied by mixing field observations and atmospheric dispersion models. The goal was to pinpoint the ^{131}I source and estimate the amount released in the environment. IRSN has developed a tool to assess an accident release into the environment when the location of the source is known.[60,61,62,63] The approach consists in the resolution of the inverse problem associated with the source-receptor relationship:[64]

$$\mu = H\sigma + \epsilon \qquad (6)$$

where μ contains air concentrations measurements; H is the source-receptor matrix calculated using atmospheric transport models, ϵ is a vector that represents errors in the system (model, instrument, and representativeness errors), and σ is the unknown temporal evolution of the release rate (source term). After all, the aim is to assess the source term σ such as the error ϵ is minimal. Considering simplified assumptions as in,[61] the resolution of the inverse problem (1) involves to minimize the following cost function:$J(\sigma) = \|\mu - H\sigma\|^2 + \lambda^2\|\sigma\|^2$

Subject to $\sigma \geq 0$

The first term of $J(\sigma)$ measures differences between observed and simulated concentrations while the second one is a regularization term, which allows avoiding unreliable solutions. The λ parameter is a scalar, which determines the magnitude of the source term fluctuations (σ). The method is applied assuming that each ^{131}I producer identified in Table 1 is a possible source location. For each producer $k, (1 \leq k \leq 6)$, the following cost function $J_k(\sigma_k)$ related to k is minimized using a quasi-Newton algorithm:

$$J_k(\sigma_k) = \|\mu - H_k\sigma_k\|^2 + \lambda^2\|\sigma_k\|^2 \qquad (7)$$

Subject to $\sigma_k \geq 0$

Then, we determine the producer k^* such as:

$$k^* = \arg \min_{1\leq k\leq 6} \{J_k(\sigma_k)\} \qquad (8)$$

This approach leads to identify the producer k^* which is the most able to reproduce the observations μ and which can be therefore considered as the most reliable source location.

Atmospheric dispersion simulations have been carried out with the Eulerian ldx model[65] developed by IRSN. Meteorological fields are provided by the ARPEGE model developed by Météo-France. The spatial resolution of the data is 0.5° × 0.5° with 3-hour time resolution. The source-receptor matrix H is computed using ldx under the approach proposed by ref.[66]. Dimensions of the computational domain are [10W, 70E], [35N, 75N] and cover the whole ^{131}I detection area. The release height is taken for the first level of the model between 0 and 40 m. For each ^{131}I producer k, daily release rates have been assessed from January 5 to February 20 (i.e. 46 days) by minimizing the cost function $J_k(\sigma_k)$. It means that the number of unknowns related to the producer k is equal to 46. In total, 196 observations were considered in the inversion process apart from the values obtained in Obninsk due to their closeness to the Karpov institute (i.e., within the same mesh cell as the source location). Even when a station did not report ^{131}I trace in the atmosphere, it contributed to constraint the inverse problem. Several values of λ in the range of $[10^{-8}, 10^{-5}]$ have been selected.

Table 3 gives the values of relative error reduction of the cost function $J_k(\sigma_k)$ obtained after minimization and the total amount of ^{131}I released between January 5 and February 20. The relative error reduction of $J_k(\sigma_k)$ is given by:

$$E_r = \frac{(J_k(\sigma_k))_{init} - (J_k(\sigma_k))_{end}}{(J_k(\sigma_k))_{init}} \times 100 \qquad (9)$$

where $(J_k(\sigma_k))_{init}$ is the initial value of the cost function $J_k(\sigma_k)$ and $(J_k(\sigma_k))_{end}$ the minimum value of $J_k(\sigma_k)$ obtained after minimization.

The maximum value of E_r is obtained when the Karpov Institute in Obninsk is assumed to be the source location. In that case, the amount of ^{131}I released estimated is ranging between 65 and 106 GBq, which is also consistent with the estimate given previously in the source apportionment section. However, these results have to be interpreted cautiously due to the inherent characteristics of the cost function $J_k(\sigma_k)$, which attributes more weight on the high values of concentrations than on the low values. Apart from the values in Obninsk, the highest concentration levels have been reported from Dubna with several tens of µBq per m^3. This station, only 200 km away from Obninsk, thus has a more significant weight than other, farther stations. Apart from that, the results on the source term are sensitive to the values of λ due to the rather small number of observations used in the inversion process. Therefore, the results given in Table 3 exemplify that the Karpov Institute (Obninsk branch) is the ^{131}I producer, since this location provides a better reproduction of the higher observed values reported in Western Russia. However, the values of $J_k(\sigma_k)$ obtained after minimization do not allow to rule on the ability to reproduce the lower levels of ^{131}I concentrations measured in Western and Central Europe.

Table 3. Relative error reduction of $\boldsymbol{J_k(\sigma_k)}$ after minimization and ^{131}I total amount estimated for each ^{131}I producer $\boldsymbol{k}$ according to the value of $\boldsymbol{\lambda}$.

^{131}I producer (location)	Released activity (GBq)	Relative error reduction (E_r)
Cis-Bio international (Saclay)	6 - 96	0 - 1%
Mallinckrodt Medical (Petten)	14 - 230	0 - 1%
Institut des RadioEléments (Fleurus)	16 - 134	0 - 1%
Polatom (Otwock-Świerk)	44 - 73	2 - 6%

Institute of Isotopes (Budapest)	41 - 256	3 - 4%
Karpov Institute, NIFKhl (Obninsk)	4 2- 78	98 - 99%

To validate the relevance of the Karpov Institute as the likely emission location, forward simulations were performed in a second stage with ldx using the source term assessed from the Karpov Institute, with $\lambda = 10^{-7}$. The comparison between simulated and observed concentrations demonstrates that the Karpov Institute is likely the origin of ^{131}I detections in Western Russia and Scandinavia in January and also in Western Russia, Poland, Germany and France during the first two weeks of February. Figure 3 illustrates the satisfactory agreement between the simulated and observed concentrations in these geographical areas. In addition, a video animation showing the hourly progress of the plume dispersion from Obninsk between January 5 and February 20, and the weekly field measurements is provided in the Supporting Information (SI).

However, the simulations are not able to reproduce the ^{131}I detections in January, located in Western Europe (France, Italy, and Germany), Hungary and Poland. Therefore, the hypothesis of concomitant releases from different facilities is a serious possibility. Indeed, the fairly high detections reported in Budapest (10 µBq m^{-3} between 23 and 30 January) may indicate that a release from the nearby Institute of Isotopes could have contributed. Forward simulations with ldx show that a release from Budapest could explain observations in Spain, France, Germany, and Czech Republic between January 16 and January 23. More advanced inverse modeling methods, based on Bayesian techniques, which do not require any knowledge of potential source locations would be a worthy addition to the present simulation results. [8, 65, 68, 69]

Figure 3

Role of meteorological conditions

The change in the detection locations mainly obeyed the prevailing wind directions over Europe in January and February 2017. The meteorological conditions had a strong impact on the detectability of the concentrations. The beginning of the year in Europe was especially characterized by poor atmospheric dispersion conditions, i.e. high-pressure fields and strong temperature gradient inversions (hereafter called “temperature inversion”) in the lower atmospheric layers, prone to increase any airborne pollutant concentration. This was the case during Weeks 3, 4, 6, and 7. Valuable information on the mixing or dispersion conditions prevailing in January 2017 over Western Europe has been derived from PM_{10} increases. Maps of peak values provided by the PREV’AIR modeling system[70] show how it affects the air quality (Fig. 4) contributing to the atmospheric pollution event.

Figure 4

Typical PM_{10} values are ranging from 10 to 20 µg m^{-3}. Simultaneous PM_{10} and ^{137}Cs peak values indicate that both airborne compounds were similarly affected by meteorological conditions as exemplified by three French locations (Fig. 5). Atmospheric detections of ^{137}Cs are typically associated with the re-suspension of contaminated soil particles.

Figure 5

Increased atmospheric activity concentrations have been also reported for other radionuclides during the ^{131}I episode, including naturally occurring ^{40}K and ^{210}Pb or anthropogenic ^{137}Cs that are detected on a routine basis by the organizations involved in the Ro5 network. Airborne concentrations peaked at their maximum levels since several months or even years. Compared with the last-6-year average values, they were higher by a factor of 1.8 for ^{40}K, 2.7 for ^{210}Pb and up to 5.2 for ^{137}Cs during the ^{131}I episode observed at the French sampling locations. No comparable increasing concentrations have been reported for ^{7}Be (factor 0.9) or ^{22}Na (factor 0.7) both of which are produced in the lower stratosphere and upper troposphere. This discrepancy points at an atmospheric process, which affects mostly radionuclides in the lower troposphere, such as ^{137}Cs or ^{131}I. Such enhancing factor becomes explainable by the temperature gradient inversion layer that could develop and last for several days, due to the radiative cooling of the atmosphere as well as low winds or lack of wind. We took an average ratio of 3.2 as representative of the average increase ratio observed at French locations for ^{137}Cs, ^{40}K, and ^{210}Pb during the time when ^{131}I was detected. Note that this ratio was almost identical for the PM_{10} increase. We called this ratio the “detection enhancing factor” that also affects particulate ^{131}I. Dividing ^{131}I concentrations by this factor leads to hypothetical ^{131}I activity concentration that would have prevailed without such enhancing meteorological conditions. In such scenario, most of the observed ^{131}I concentrations would have fallen below usual detection limits. This is sufficient to explain why the Ro5 network does not measure ^{131}I on a regular basis, even if it can be hypothesized that ^{131}I will become present more or less routinely at ultra-traces levels as the result of sporadic release events or routine releases from the booming radiopharmaceutical industry.

Detection of harmless traces of airborne ^{131}I in January/February 2017 in Europe resulted from a combination of multiple sources and poor atmospheric dispersion conditions. The main releases capable to imprint trace concentrations on a wide scale belong to the radiopharmaceutical industry that remains the most probable source in the present case study. Based on field observations and inverse dispersion modeling, the Obninsk region appears as the most likely involved emission area, but with a release amount consistent with usual authorized limits for the Karpov Institute. This makes this unusual ^{131}I episode over Europe the first case of a regular/routine release of a radionuclide resulting in continental detections by members of the Ro5 monitoring network. Other releases from radiopharmaceutical production units located in Hungary and to a lesser extent in Poland may explain the concentrations observed on Week 2 and Week 4, respectively on a local to regional scale. Apart from radiopharmaceutical release sources, there were multiple secondary additional emissions (e.g. from nuclear medicine hospitals or sewage sludge incinerators) that were too weak to significantly contribute to European-wide detections when considering them alone, but that may have exhibited a dominant contribution on a local scale. In any case, nuclear power plant-derived releases of ^{131}I appear too weak to be detected even by monitoring stations located in their vicinity. All of those detections would probably have gone unnoticed without unusually disadvantageous weather conditions marked by strong inversions of the vertical temperature gradient in the lower atmospheric layers. As a result, they led to concentrations exceeding the usual detection limits in use within the Ro5 network. While likely to be dominant, the lack of reports of the gaseous fraction of ^{131}I also denotes the demand to improve both its detection limits and the number of samplers for gaseous radioiodine. Finally this event also confirms the capability of the organizations involved in the Ro5 informal network, and in charge of monitoring airborne radioactivity to detect unexpected radionuclides or unusual concentrations, down to the 0.1 – 1 μBq m^{-3} range, thus ensuring a highly sensitive monitoring even at concentration levels of no concern for human health.

Associated content

Supporting Information: Video animation of the plume dispersion from Obninsk.

Author information

(O.M.) Phone: +33-4-42-19-9608; Email: olivier.masson@irsn.fr

Acknowledgements

We wish to express our deep thanks to the following organizations who also shared their data and comments: University of the Basque Country (UPV/EHU), Bilbao, Spain; Federal Office of Public Health (FOPH/OFSP), Bern, Switzerland; National Institute of Public Health and the Environment (RIVM), Bilthoven, The Netherlands; Swedish Defence Research Agency (FOI) and Swedish Radiation Protection Agency (SSM), Stockholm, Sweden; DTU Nutech, Center for Nuclear Technologies (DTU), DK - Roskilde, Denmark; Icelandic Radiation Safety Authority (IRSA), Reykjavik, Iceland; and the Comprehensive nuclear Test Ban Treaty Organization (CTBTO) to giving access to their vDEC data bank (https://www.ctbto.org/specials/vdec/) and permission to use their data for the interpretation presented herein.

Disclaimer

The views expressed in this study are those of the authors and do not necessarily reflect the views of the CTBTO Preparatory Commission.

References

1. Masson, O.; Baeza, A.; Bieringer, J.; Brudecki, K.; Bucci, S.; Cappai, M.; Carvalho, F. P.; Connan, O.; Cosma, C.; Dalheimer, A.; Didier, D.; Depuydt, G.; De Geer, L. E.; De Vismes, A.; Gini, L.; Groppi, F.; Gudnason, K.; Gurriaran, R.; Hainz, D.; Halldorsson, O.; Hammond, D.; Hanley, O.; Holey, K.; Homoki, Z.; Ioannidou, A.; Isajenko, K.; Jankovic, M.; Katzlberger, C.; Kettunen, M.; Kierepko, R.; Kontro, R.; Kwakman, P. J. M.; Lecomte, M.; Leon Vintro, L.; Leppanen, A. P.; Lind, B.; Lujaniene, G.; McGinnity, P.; McMahon, C.; Mala, H.; Manenti, S.; Manolopoulou, M.; Mattila, A.; Mauring, A.; Mietelski, J. W.; Moller, B.; Nielsen, S. P.; Nikolic, J.; Overwater, R. M. W.; Palsson, S. E.; Papastefanou, C.; Penev, I.; Pham, M. K.; Povinec, P. P.; Rameback, H.; Reis, M. C.; Ringer, W.; Rodriguez, A.; Rulik, P.; Saey, P. R. J.; Samsonov, V.; Schlosser, C.; Sgorbati, G.;

Silobritiene, B. V.; Soderstrom, C.; Sogni, R.; Solier, L.; Sonck, M.; Steinhauser, G.; Steinkopff, T.; Steinmann, P.; Stoulos, S.; Sykora, I.; Todorovic, D.; Tooloutalaie, N.; Tositti, L.; Tschiersch, J.; Ugron, A.; Vagena, E.; Vargas, A.; Wershofen, H.; Zhukova, O. Tracking of Airborne Radionuclides from the Damaged Fukushima Dai-Ichi Nuclear Reactors by European Networks. *Environ. Sci. Technol.* **2011,** *45*, (18), 7670-7677.

2. Uematsu, M.; Merrill, J. T.; Patterson, T. L.; Duce, R. A.; Prospero, J. M. Aerosol residence times and iodine gas/particle conversion over the North Pacific as determined from Chernobyl radioactivity. *Geochem. J.* **1988,** *22*, 157-63.

3. Petrova, K.; Jankovec M.; Fojtíkova I.; Hůlka J. New Challenges in Crisis Communication – the Results of Sociological Survey in the Czech Republic. Proceedings oft he RICOMET 2017 conference: Social and ethical aspects of decision-making in radiological risk situations. 27th-29th June, **2017** Vienna, Austria . IAEA

4. Detection of radioactive iodine at trace levels in Europe in January **2017**. http://www.irsn.fr/en/newsroom/news/pages/20170213_detection-of-radioactive-iodine-at-trace-levels-in-europe-in-january-2017.aspx (accessed September 29, 2017).

5. Artus, J. C.; Faurous, P. Atmospheric and liquid releases by hospitals. *Radioprotection* **1994,** *29*, (4), 489-501.

6. Matthews, M.; Vandergrift, G.; Ponsard, B.; Solin, L. M.; Dolinar, G.; Carranza, E.; Ringbom, A.; Hoffman, E.; Duran, E.; Turinetti, J.; Achim, P.; Hebel, S.; Saey, P.; Ramamoorthy, N.; Mikolajczak, R.; Fisher, D.; Higgy, R.; Biegalski, S.; Sameh, A. A.; Barbosa, L.; Zähringer, M.; Mercer, D.; Popov, V.; Becker, A.; Bowyer, T.; Cutler, C.; Tsipenyuk, Y. M.; Ungar, K.; Schraick, I.; Deconnick, B.; Amaya, D.; Camps, J.; Rao, A.; Auer, M.; Steinhauser, G.; Solomon, S. **2010,** *Workshop on Signatures of Medical and Industrial Isotope Production—A Review, PNNL-19294*. PNNL: Richland.

7. Leelössy, Á.; Mészáros, R.; Kovács, A.; Lagzi, I.; Kovács, T. Numerical simulations of atmospheric dispersion of iodine-131 by different models. *PLoS ONE,* **2017,** *12* (2): e0172312. DOI:10.1371/journal.pone.0172312.

8. Tichý, O.; Šmídl, V.; Hofman, R.; Šindelářová, K.; Hýža, M.; Stohl, A. Bayesian inverse modeling and source location of an unintended I-131 release in Europe in the fall of 2011. *Atmos. Chem. Phys. Discuss.* **2017,** 1-24.

9. Tombuyses, B.; Schmitz, F.; Wertelaers, A.; Schrauben, M.; Van Den Berghe, Y.; Drymael, H. Incidental release in IRE. https://www.eurosafe-forum.org/sites/default/files/Presentations2009/Seminar5/Slides/5.2-Incidental%20release%20in%20IRE.pdf

10. Vandecasteele, C. M.; Sonck, M.; Degueldre, D. Rejet accidentel d'iode-131 par l'IRE sur le site de Fleurus : Retour d'expérience de l'autorité de sûreté belge. *Radioprotection* **2011,** *46*, (2), 159-173 (in French).

11. International Union of Radioecology (IUR). The Ring of Five task group. http://www.iur-uir.org/en/task-groups/id-22--ring-of-five-task-group (accessed September 29, 2017).

12. Bieringer, J.; Bleher, M.; Dalheimer, A.; Dersch, G.; Herrmann, J.; Peter, J.; Roos, N.; Steinkopff, T.; Tait, D.; Wershofen, H. Messergebnisse aus Deutschland in Zusammenhang mit den Reaktorunfällen in Fukushima (in German). In *Umweltradioaktivität in der Bundesrepublik Deutschland - Stand 2011*, Trugenberger-Schnabel, A.; Peter, J.; Kanzliwius, R.; Hachenberger, C.; Bernhard-Stör, C., Eds. Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU): Bonn, 2012.

13. Steinhauser, G.; Merz, S.; Kübber-Heiss, A.; Katzlberger, C. Using animal thyroids as ultra-sensitive biomonitors for environmental radioiodine. *Environ. Sci. Technol.* **2012,** *46*, (23), 12890-12894.

14. IAEA Source of Iodine-131 in Europe Identified. http://www.iaea.org/newscenter/pressreleases/2011/prn201127.html (July 2012).

15. http://www.stuk.fi/web/en/topics/environmental-radiation/radioactivity-in-outdoor-air

16. http://egasmro.ru/files/documents/ro_bulletins/byulleten_rorf_09_2017.pdf, p 4.

17. International Monitoring System. http://ims.ctbto.org/(accessed June 30, 2017).

18. Map of monitoring stations https://www.ctbto.org/map/(accessed June 2017).

19. Matthews, K.M.; Bowyer, T.W.; Saey, P.R.J.; Payne, R.F. The Workshop on Signatures of Medical and Industrial Isotope Production - WOSMIP; Strassoldo, Italy, 1-3 July 2009. *J. Environ. Radioact.* **2012**, 110, 1-6.

20. Lee, S.-K.; Beyer, G.J.; Lee, J.S. Development of industrial-scale fission ^{99}Mo production process using low enriched uranium target. *Nucl. Engineering and Technol.* **2016***,* 48, 613-623.

21. Karpov Institute of Physical Chemistry Activity Report. http://www.rosatom.ru/upload/iblock/334/334f8a20cd4f2b02cb25e58b48190bd8.pdf (in Russian) (May 2017).

22. Zhuikov, B. L. Production of medical radionuclides in Russia: Status and future—a review. *Appl. Radiat. Isot.* **2014,** *84*, 48-56.

23. Ageeva, N. V.; Kim, V. M.; Vasilieva, K. I.; Katkova, M. N.; Volokitin, A. A.; Polyanskaya, O. N. Long-term monitoring airborne I-131 in the surface layer in Obninsk city, Kaluga region. *Radiation & Risk* **2015,** *24*, (1), 96-107.

24. http://www.bip.paa.gov.pl/paa/obiekty-jadrowe/roczne-oceny-stanu-bezp/6162,Roczne-oceny-stanu-bezpieczenstwa-obiektow-jadrowych.html. (in Polish)

25. https://www.rvo.nl/ (in Dutch)

26. http://www.cea.fr/Documents/Rapport-TSN-Saclay-2013.pdf

27. https://ec.europa.eu/energy/sites/ener/files/documents/tech_report_hungary_2012_en.pdf

28. http://www.rpatyphoon.ru/upload/medialibrary/fef/byulleten_rorf_01_2017.pdf

29. http://www.rpatyphoon.ru/upload/medialibrary/e88/byulleten_rorf_02_2017.pdf

30. Kitto, M. E.; Fielman, E. M.; Fielman, S. E.; Gillen, E. A. Airborne ^{131}I at a background monitoring site. *J. Environ. Radioact.* **2005**, 83, 129-136.

31. IAEA Radiopharmaceuticals: Production and Availability. https://www.iaea.org/About/Policy/GC/GC51/GC51InfDocuments/English/gc51inf-3-att2_en.pdf (May 2017).

32. Jiménez, F.; Debán, L.; Pardo, R.; López, R.; García-Talavera, M. Levels of ^{131}I and six natural radionuclides in sludge from the sewage treatment plant of Valladolid, Spain. *Water Air Soil Pollut.* **2011,** *217*, (1), 515-521.

33. Fischer, H. W.; Ulbrich, S.; Pittauerová, D.; Hettwig, B. Medical radioisotopes in the environment - following the pathway from patient to river sediment. *J. Environ. Radioact.* **2009**, *100*, (12), 1079-1085.

34. Strahlenschutzkommission (SSK), *Emittlung der Vorbelastung durch Radionuklid-Ausscheidungen von Patienten in der Nuklearmedizin. Empfehlungen der Strahlenschutzkommission (in German)*. SSK: Bonn, **2004**. (in German).

35. StrlSchV, Verordnung über den Schutz vor Schäden durch ionisierende Strahlen (Strahlenschutzverordnung - StrlSchV). *BGBl. I* **2001**, 1714; 2002 I S. 1459 (German Federal Law).

36. Euratom, Council Directive 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation. *Official Journal of the European Union* **2013,** *L 13/1*, 7.1.2014.

37. Hormann, V.; Fischer, H. W. The physicochemical distribution of ^{131}I in a municipal wastewater treatment plant. *Internal Report, University of Bremen* **2017**, 1-17.

38. BMUB, *Umweltradioaktivität und Strahlenbelastung, Jahresbericht 2014*. Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety: Bonn (Germany), 2015.

39. BMUB, *Umweltradioaktivität und Strahlenbelastung, Jahresbericht 2015*. Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety: Bonn (Germany), 2016.

40. FWR - Foundation for Water Research, *Sewage sludge: Operational and Environmental Issues, FR/R0001*. 4th ed.; FWR: Buck, UK, 2016.

41. Bień, J. D. Zagospodarowanie komunalnych osadów ściekowych metodami termicznymi (Utilisation of Sewage Sludge in Poland by Thermal Methods) (in Polish). *Inżynieria I Ochrona Środowiska* **2015,** *15*, (4), 439-449.

42. Sroda, K.; Kijo-Kleczkowska, A. Analysis of combustion process of sewage sludge in reference to coals and biomass. *Arch. Min. Sci.* **2016,** *61*, (2), 425-442.

43. Statistisches Bundesamt, *Statistisches Jahrbuch 2013 (in German)*. Statistisches Bundesamt: Wiesbaden, 2013.

44. Statistisches Bundesamt, *Statistisches Jahrbuch 2014 (in German)*. Statistisches Bundesamt: Wiesbaden, 2014.

45. Wiechmann, B.; Dienemann, C.; Kabbe, C.; Brandt, S.; Vogel, I.; Roskosch, A. *Sewage sludge management in Germany*. Umweltbundesamt: Bonn, 2013.

46. Bundesamt für Strahlenschutz. http://www.bfs.de/SharedDocs/Bilder/BfS/DE/ion/umwelt/fukushima-spurenmessstellen-jod.jpg (May 2016).

47. Malmqvist, L.; Kristiansson, K. Experimental evidence for an ascending microflow of geogas in the ground. *Earth Planet. Sci. Letters*, **1984,** *70*, 407-416.

48. Sakoda, A.; Ishimori, Y.; Yamaoka K. A comprehensive review of radon emanation measurements for mineral, rock, soil,mill tailing and fly ash. *Appl. Radiat. Isot.*, **2011**, *69*, 1422–1435.

49. Shultis, J. K.; Faw, R. E. *Fundamentals of Nuclear Science and Engineering*. CRC Press: Boca Raton, 2008.

50. Taylor, S. R.; McLennan, S. M. *The continental crust: its composition and evolution*. Blackwell Scientific Publications: Oxford, 1985.

51. Andrews, J. N.; Davis, S. N.; Fabryka-Martin, J.; Fontes, J. C.; Lehmann, B. E.; Loosli, H. H.; Michelot, J. L.; Moser, H.; Smith, B.; Wolf, M. The in situ production of radioisotopes in rock matrices with particular reference to the Stripa granite. *Geochim. Cosmochim. Acta* **1989,** *53*, (8), 1803-1815.

52. Fabryka-Martin, J. T.; Davis, S. N.; Elmore, D.; Kubik, P. W. In situ production and migration of ^{129}I in the Stripa granite, Sweden. *Geochim. Cosmochim. Acta* **1989,** *53*, (8), 1817-1823.

53. Santschi, P. H.; Xu, C.; Zhang, S.; Schwehr, K. A.; Grandbois, R.; Kaplan, D. I.; Yeager, C. M. Iodine and plutonium association with natural organic matter: A review of recent advances. *Appl. Geochem.* **2017,** 85, 121-127. DOI: 10.1016/j.apgeochem.2016.11.009.

54. Ashworth, D. J. Transfers of Iodine in the Soil–Plant–Air System: Solid–Liquid Partitioning, Migration, Plant Uptake and Volatilization. In *Comprehensive Handbook of Iodine*, Preedy, V. R.; Burrow, G. N.; Watson, R. R., Eds. Academic Press: Oxford, 2009.

55. Kalinowski, M. B. Characterization of prompt and delayed atmospheric radioactivity releases from underground nuclear tests at Nevada as a function of release time. *J. Environ. Radioact.* **2011,** *102*, 824-836.

56 Bostock, A. C.; Shaw, G.; Bell, J. N. B. The volatilisation and sorption of ^{129}I in coniferous forest, grassland and frozen soils. *J. Environ. Radioact.* **2003,** *70*, (1–2), 29-42.

57. Holden Norman, E.; Hoffman Darleane, C. Spontaneous fission half-lives for ground-state nuclide (Technical report). In *Pure and Applied Chemistry*, 2000; Vol. 72, p 1525.

58. Sabu, D. D. On mass-yield of xenon and krypton isotopes in the spontaneous fission of uranium. *J. Inorg. Nucl. Chem.* **1971,** *33*, (5), 1509-1513.

59. Ashizawa, F. T.; Kuroda, P. K. The occurrence of the short-lived iodine isotopes in natural and in depleted uranium salts. *J. Inorg. Nucl. Chem.* **1957,** *5*, (1), 12-22.

60. Saunier, O.; Mathieu, A.; Didier, D.; Tombette, M.; Quélo, D.; Winiarek, V.; Bocquet, M. An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations, *Atmos. Chem. Phys.*, **2013**, 13, 11403–11421, DOI:10.5194/acp-13-11403-2013.

61. Saunier, O.; Mathieu, A.; Sekiyama, T.T.; Kajino, M.; Adachi, K.; Bocquet, M.; Igarashi, Y.; Maki, T.; Didier, D. A new perspective on the Fukushima releases brought by newly available ^{137}Cs air concentration observations and reliable meteorological fields. HARMO 2016 - 17th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, Proceedings. **2016**, Volume 2016-May, , 528-535.

62. Winiarek, V.; Bocquet, M.; Saunier, O.; Mathieu, A. Estimation of errors in the inverse modeling of accidental release of atmospheric pollutant: Application to the reconstruction of the cesium-137 and iodine-131 STs from the Fukushima Daiichi power plant. *J. Geophys. Res.*, **2012**, *117*, D05122.

63. Winiarek, V.; Bocquet, M.; Duhanyan, N.; Roustan, Y.; Saunier, O.; Mathieu, A. Estimation of the caesium-137 source term from the Fukushima Daiichi nuclear power plant using a consistent joint assimilation of air concentration and deposition observations, *Atmos. Environ.*, **2014**, *82*, 268–279.

64. Seibert, P.; Frank, A. Source-receptor matrix calculation with a Lagrangian particle dispersion model in backward mode. *Atmos. Chem. Phys.*, **2004**, 4, 51–63.

65. Quelo, D.; Krysta, M.; Bocquet, M.; Isnard, O.; Minier, Y.; Sportisse, B. Validation of the Polyphemus platform on the ETEX, Chernobyl and Algeciras cases, *Atmos. Environ.*, **2007**, 41, 5300-5315.

66. Winiarek, V.; Vira, J.; Bocquet, M.; Sofiev, M.; Saunier, O. Towards the operational estimation of a radiological plume using data assimilation after a radiological accidental atmospheric release, *Atmos. Environ.*, **2011**, 45, 2944-2955.

67. Delle Monache, L.; Lundquist, J. K.; Kosovic, B.; Johannesson, G.; Dyer, K. M.; Aines, R. D.; Chow, F. K.; Belles, R. D.; Hanley, W. G.; Larsen, S. C.; Loosmore, G. A.; Nitao, J. J.; Sugiyama, G. A.; Vogt, P. J. Bayesian inference and Markov chain Monte Carlo sampling to reconstruct a contaminant source on a continental scale. *J. Applied Meteo. and Climat.*, **2008**, 47, 2600–2613.

68. Liu, Y.; Haussaire, J.-M.; Bocquet, M.; Roustan, Y.; Saunier, O.; Mathieu, A. Uncertainty quantification of pollutant source retrieval: comparison of Bayesian methods with application to the Chernobyl and Fukushima Daiichi accidental releases of radionuclides: uncertainty Quantification of Pollutant Source Retrieval. *Q. J. R. Meteorol. Soc.,* **2017**, 143, 2886–2901.

69. Yee, E.; Lien, F.-S.; Keats, A.; D'Amours, R. Bayesian inversion of concentration data: source reconstruction in the adjoint representation of atmospheric diffusion, *J. Wind Engin. Indus. Aerodyn*., **2008**, 96, no. 10-11, 1805–1816.

70. PREV'AIR www.prevair.org (April 2017).

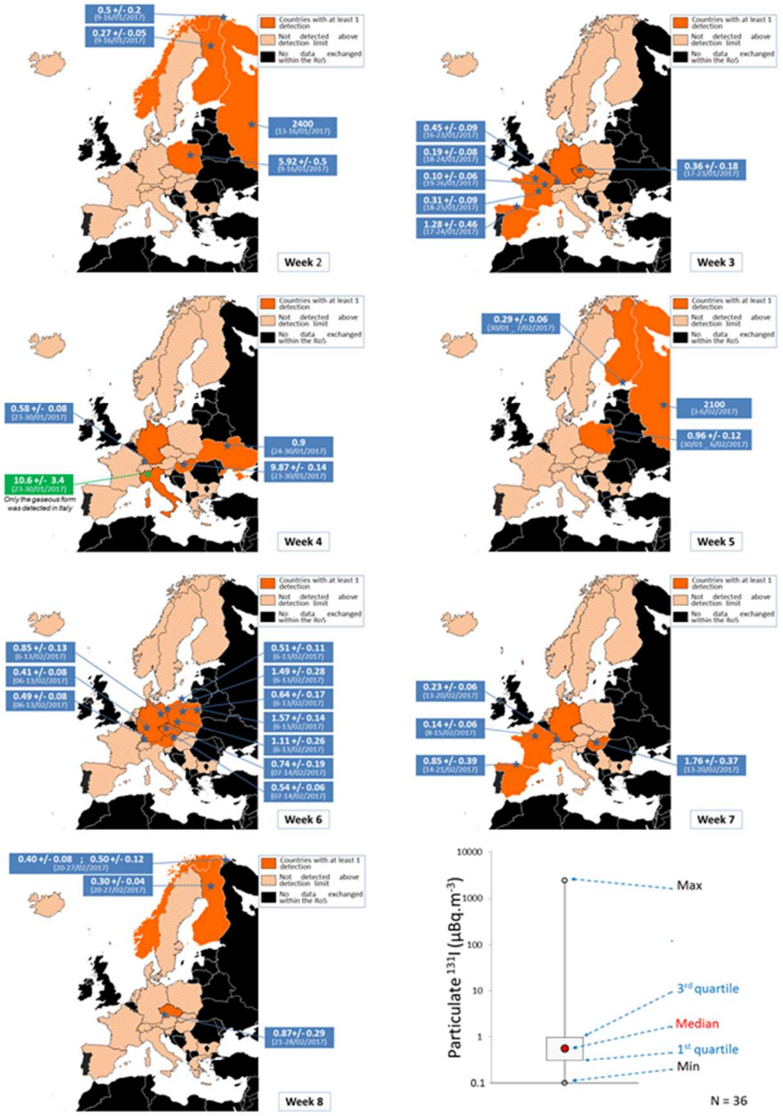


Figure 1. Weekly changes of particulate ^{131}I (µBq m^{-3}) in January / February 2017 and box-and-whisker plot of the 36 weekly values. CTBTO data are not presented since based on daily samples.

190x254mm (96 x 96 DPI)

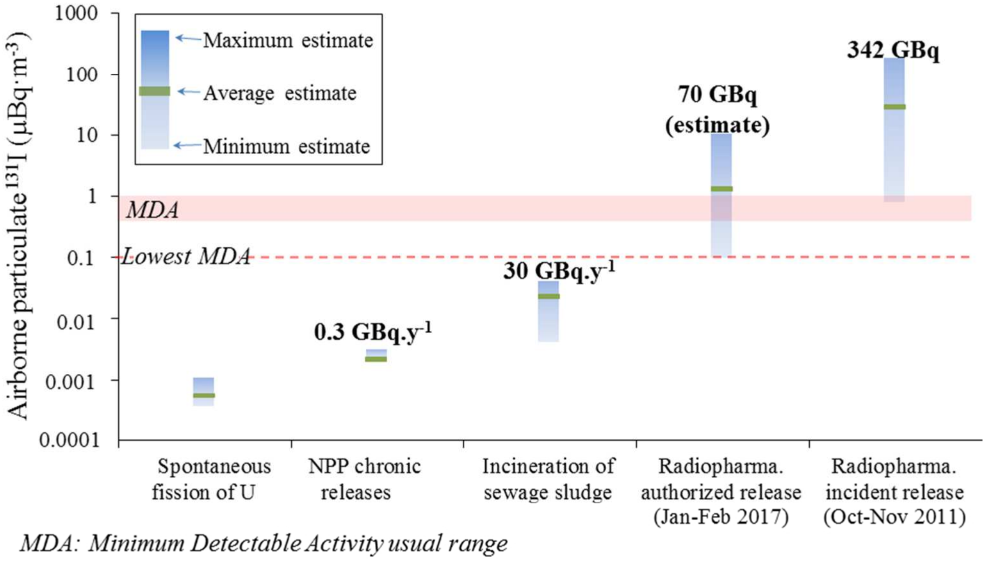


Figure 2. Source apportionment of particulate ^{131}I in the air over Europe in January/February 2017 based on source term estimates.

254x190mm (96 x 96 DPI)

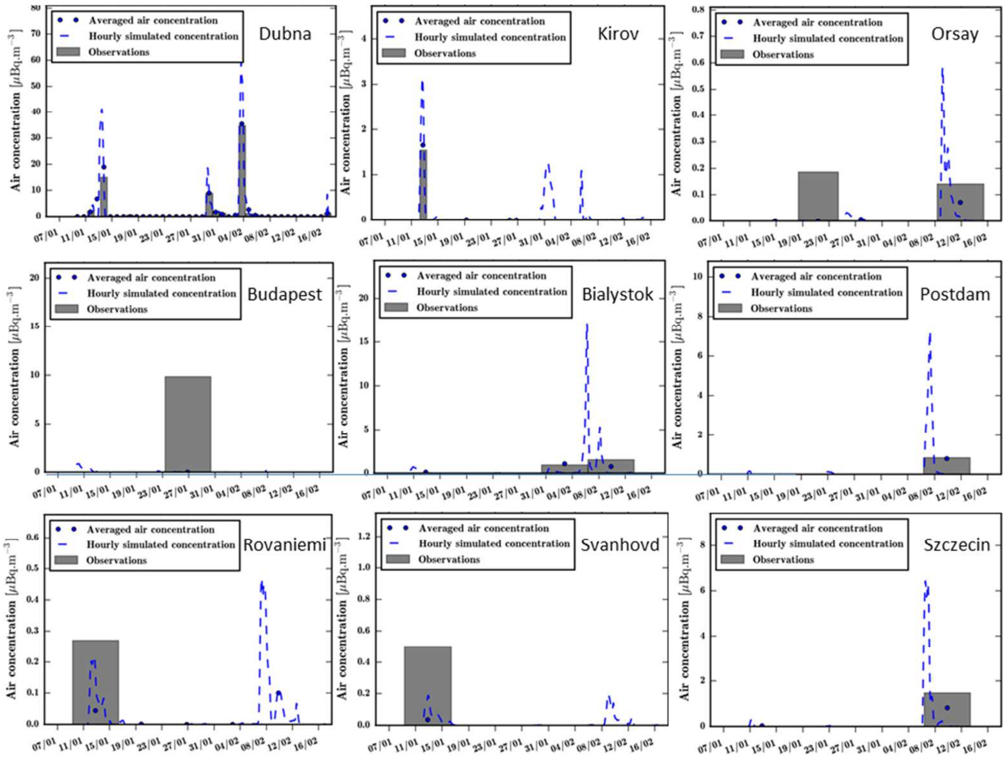
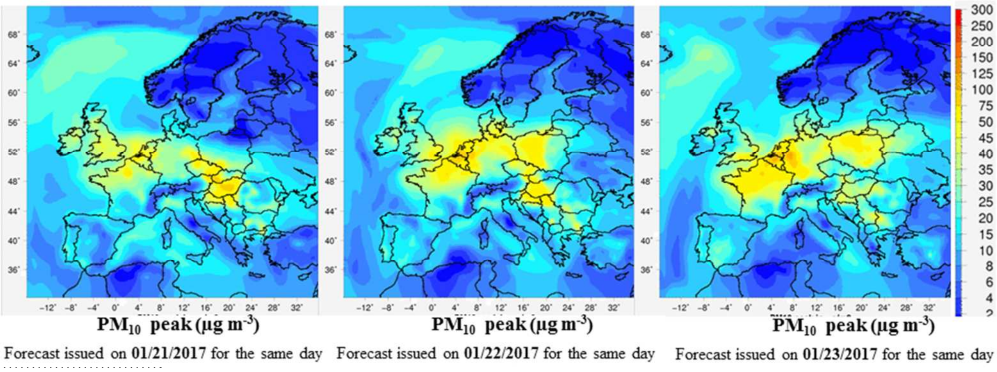


Figure 3. Observed ^{131}I activity concentrations (grey blocks), and simulated activity concentrations derived from the source term assessed for the Karpov Institute. Blue dashes are hourly simulated concentrations and blue circles are centered on the respective time interval measurements.

254x190mm (96 x 96 DPI)



PM_{10} peak ($\mu g\ m^{-3}$)
Forecast issued on **01/21/2017** for the same day

PM_{10} peak ($\mu g\ m^{-3}$)
Forecast issued on **01/22/2017** for the same day

PM_{10} peak ($\mu g\ m^{-3}$)
Forecast issued on **01/23/2017** for the same day

Figure 4. PM10 peak value forecasts (µg m^{-3}) over Europe, 21-23 January 2017. The maps are representative for large-scale phenomena; they cannot reproduce local aspects of air pollution.

254x190mm (96 x 96 DPI)

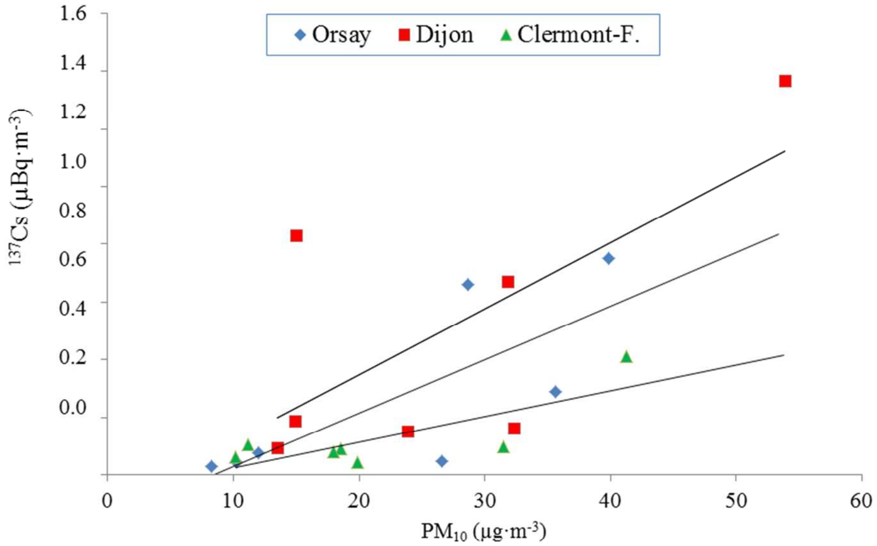


Figure 5. Example of weekly PM10 and ^{137}Cs at French sampling locations having ^{131}I detections in January and February 2017.

254x190mm (96 x 96 DPI)

254x190mm (96 x 96 DPI)