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Global risk of radioactive fallout after nuclear reactor accidents

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Abstract

Reactor core meltdowns of nuclear power plants are rare, yet the consequences are catastrophic. But what is meant by “rare”? And what can be learned from the Chernobyl and Fukushima incidents? Here we assess the risk of exposure to radioactivity due to atmospheric dispersion of gases and particles following severe nuclear accidents, using particulate ^{137}Cs and gaseous ^{131}I as proxies for the fallout. It appears that previously the occurrence of major accidents and the risks of radioactive contamination have been underestimated. Using a global model of the atmosphere we compute that on average, in the event of a core melt of any nuclear power plant worldwide, more than 90 % of emitted ^{137}Cs would be transported beyond 50km and about 50 % beyond 1000 km distance. This corroborates that such accidents have large-scale and trans-boundary impacts. Although the emission strengths and atmospheric removal processes of ^{137}Cs and ^{131}I are quite different, the radioactive contamination patterns over land and the human deposition exposure are computed to be similar. High human exposure risks occur around reactors in densely populated regions, notably in southern Asia where a core melt can subject 55 million people to radioactive contamination. The recent decision by Germany to phase out its nuclear reactors will reduce the national risk, though a large risk will still remain from the reactors in neighbouring countries.

1 Introduction

An accident risk assessment of nuclear power plants (NPPs) by the US Nuclear Regulatory Commission in 1975 estimated the probability of a core melt at 1 in 20 000 per year for a single reactor unit (NRC, 1975). A follow-up report in 1990 adjusted this number and indicated that the core damage frequency is not a value that can be calculated with certainty, though an appendix presented the following likelihood of a catastrophic accident (NRC, 1990):

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- a. Probability of core melt 1 in 10 000 per year.
- b. Probability of containment failure 1 in 100.
- c. Probability of unfavourable wind direction 1 in 10.
- d. Probability of meteorological inversion 1 in 10.
- e. Probability of evacuation failure 1 in 10.

The product of these possibilities is 1 in 1 billion per year for a single reactor (note that this assumes that factors (a)–(e) are independent, which is not the case, so that the actual risk of a catastrophic accident should be higher than this). Given this, with a total of 440 active reactors worldwide (IAEA, 2011; Supplement), and an estimated mean remaining lifetime of 20–25 yr (together ~10 000 reactor years), then the probability of such a major accident occurring in this period would be roughly 1 in 100 000. In light of the uncertainties, the simplicity of this calculation is appealing.

However, based on the evidence over the past decades it appears that the probabilities (a) and (b) have been substantially underestimated. Furthermore, by using a state-of-the-art global atmospheric model we can directly compute the anticipated dispersion of radionuclides, avoiding the need to guess the factors (c) and (d). In doing so, we find that the vast majority of the radioactivity is transported outside an area of 50 km radius, which can undermine evacuation measures, especially if concentrated deposition occurs at much greater distances from the accident, as was the case for Chernobyl in May 1986. Furthermore, even if an evacuation is successful in terms of saving human lives, large areas around the reactors are made uninhabitable for decades afterwards. Therefore, we argue that such events are catastrophic irrespective of evacuation failure or success, and exclude the factor (e).

To gain an overview of the geographical and population-weighted risk of contamination, we present calculations of the amount of radioactive fallout expected to occur within various distances around each reactor. We focus on the most severe NPP disasters, defined as level 7 major accidents on the International Nuclear Event Scale

(INES). Since we cannot predict the radioactivity that would actually be released by future events, which depends on many factors such as reactor type, fuel and capacity, we focus on the relative amount that would occur for any core melt. To at least provide an approximation of the absolute amount of radioactivity released, we use analyses of the well-documented Chernobyl reactor accident as a proxy (Smith and Beresford, 2005; IAEA, 2006). In the discussion we also address preliminary estimates of the Fukushima emissions and their implications for our risk assessment.

2 The Chernobyl accident

Radioactivity from the Chernobyl nuclear reactor incident, which occurred on 26 April 1986, impacted the entire Northern Hemisphere. Anspaugh et al. (1988) estimated that the collective dose to humans was about 930 000 Gray (the dose of ionizing radiation, expressed by 1 Gy, is equivalent to 1 Sievert (Sv), which represents the absorption of 1 Joule by one kg of matter). Nuclear accidents release both gaseous and particulate radioisotopes. The total radionuclide release of Chernobyl was influenced by the ignition of the graphite-mediated reactor, and amounted to >12 000 PBq (IAEA, 2006) (P is peta = 10^{15}) (Table 1). The on-going fires released a large amount of fuel particles (“hot” particles) carrying isotopes of cerium, zirconium, molybdenum, neptunium and plutonium (Smith and Beresford, 2005; IAEA, 2006). These particles were relatively large and quickly sedimented from the atmosphere, primarily contaminating an area of about 30km around the reactor.

Since the Chernobyl type reactor technology is now considered obsolete, we rather focus on the radionuclides that were emitted as gases and attached to ambient aerosol particles, e.g. the semi-volatile isotopes of iodine, strontium, caesium, tellurium, ruthenium and barium ($^{131,133}\text{I}$, $^{89,90}\text{Sr}$, $^{134,137}\text{Cs}$, ^{132}Te , $^{103,106}\text{Ru}$ and ^{140}Ba). These radionuclides were mostly found on small particles with a radius of $r \leq 1 \mu\text{m}$, which deposit slowly by gravitational settling and are more effectively removed by rainfall, usually further downwind than the large particles.

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Although only a small fraction of the radionuclides from the Chernobyl core melt was released as ^{137}Cs , i.e. 85 PBq (about 27 kg), ^{137}Cs (half-life of 30 yr) is used to map the deposition because it is straightforward to measure and is radiologically important on a long time scale (Smith and Beresford, 2005; IAEA, 2006). ^{131}I (half-life of 8 days) is also important, especially in the first weeks after an accident, as it is released in relatively large quantities and can rapidly enter the food chain (IAEA, 2006; WHO, 2006; Christodouleas et al., 2011). To put the emissions by Chernobyl into perspective, we list in Table 1 the known level 4 to 7 accidents and their estimated radioactivity release to the atmosphere, largely based on non-official information gathered from the Internet.

Since there is little reliable information besides Chernobyl about the release and even less about the deposition of radionuclides from a catastrophic accident, we apply the Chernobyl data in our model to give a first approximation, and simulate that a core melt of each reactor releases 85 PBq of ^{137}Cs and 1760 PBq of ^{131}I over a period of one year. The duration of the emission period is not decisive for these calculations since the total deposition onto the ground is the most relevant parameter for a risk assessment, and by integrating over a year we capture the annual range of meteorological conditions, thus providing an average over the different seasons.

3 Global model calculations

Simulations of particulate ^{137}Cs and gaseous ^{131}I have been performed with the ECHAM5/MESSy Atmospheric Chemistry (EMAC) general circulation model (Roeckner et al., 2006; Jöckel et al., 2006; Kerkweg et al., 2006, 2007; Tost et al., 2007; Burrows et al., 2009; Pringle et al., 2010). We applied it at T106 horizontal resolution (corresponding to a quadratic Gaussian grid of about $1.1^\circ \times 1.1^\circ$ in longitude and latitude, approximately 100×100 km) and 31 vertical layers up to 10 hPa. The applied sub-models, which simulate all relevant meteorological processes for the dispersion of the aerosols, are: CLOUD, CONVECT, RAD4ALL, PTRAC, CVTRANS, ONLEM,

DRYDEP, SEDI, SCAV (for a description, see <http://www.messy-interface.org/>). We simulated the year 2005, which we consider representative, i.e. without strong annual modes (e.g. El Niño, North Atlantic Oscillation), by using meteorological analyses from the European Centre for Medium-range Forecasts (ECMWF) to nudge the general circulation model (Lelieveld et al., 2007). The mean aerosol lifetime of ^{137}Cs in the atmosphere, about one week, is much shorter than the decay time of the radionuclei; therefore the removal of particulate radioactivity is controlled by wet and dry deposition of the particles, rather than atmospheric decay. ^{131}I has a much shorter half-life, so that a significant amount can decay radioactively before depositional loss; we assume that it travels in the gas phase as $^{131}\text{I}_2$ and have represented its dry deposition using the method of Ganzeveld et al. (1998), based on the concept of Wesely (1989). We apply a Henry's law coefficient of $3.1 \text{ mol l}^{-1} \text{ atm}^{-1}$, indicating a low solubility. For a review of deposition modelling methods we refer to Sportisse (2007).

In the model we emit the ^{137}Cs on aerosol particles with a mean radius of $0.5 \mu\text{m}$ by introducing them into the surface layer of about 60m depth. We assume that ^{131}I and ^{137}Cs are released gradually and not explosively or by large fires, which would increase the emission height and generally enhance the long distance transport. Since the chemical composition of the particles is not known, we adopted two categories, one "soluble" and one "insoluble", which represent the extremes of possible particle characteristics. The main difference between the two is that only soluble particles are removed by nucleation scavenging, i.e. they can act as cloud condensation nuclei (CCN), though do not exert feedbacks on cloud formation processes. Both soluble and insoluble particles are removed by impaction scavenging (Tost et al., 2007), aerosol sedimentation, and dry deposition (Kerkweg et al., 2006). Thus, the insoluble particles have a longer lifetime due to reduced wet deposition and the consequently extended transport in the atmosphere. The difference between the results from the two simulations is typically less than 30 %.

To test the above assumptions and the model setup, we simulated the Chernobyl core melt in 1986 to study the distribution of ^{137}Cs deposition and compare it

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with a compilation-interpolation of measurement data presented by Smith and Beresford (2005), based on the original work of De Cort et al. (1998). Since the deposition data are not publicly available, we evaluated the model output by comparing with a deposition map based on these data (Smith and Beresford, 2005; Peplow, 2006), applying the same colour scaling in Fig. 1. We find that that our results agree well e.g. with the map of Peplow (2006) (see also IAEA, 2006), especially of our soluble tracer, indicating ^{137}Cs deposition in excess of 40 kBq m^{-2} in the Ukraine, Scandinavia, Northern Greece, southern Germany and Austria (Fig. 1).

This qualitative agreement is quite satisfactory, especially because quantitative agreement cannot be expected due to the neglect of the large particles emitted by Chernobyl fires and the difficulty of accurately simulating the emission time profile. Considering that we achieve the best agreement with the soluble tracer, and since the majority of particles acts as CCN (Pruppacher and Klett, 1997), we henceforth assume that 75 % of the aerosol is removed by nucleation scavenging (i.e. 75 % soluble and 25 % insoluble particles). Our total ^{137}Cs deposition distribution also compares favourably with the high-resolution regional model results of Brandt et al. (2002), notably the map for which the most sophisticated deposition method was applied (their Fig. 7).

Subsequently we emitted gaseous ^{131}I and particulate ^{137}Cs from each of the 440 active reactors worldwide for the year 2005 (the reactors, locations and operational lifetimes are listed in the Supplement). The calculations indicate that on average only about 8 % of the ^{137}Cs deposits within 50 km of the source, ~30 % within 500 km, ~50 % within 1000 km, and that about 25 % is transported beyond 2000 km. Clearly, radionuclides from a reactor core melt anywhere in the world would not only be deposited locally. The variability of these fractions for different regions, e.g. in Europe, the USA and Asia is relatively small, even though the retention of emissions near the surface in temperate and high latitudes is typically stronger than in the tropics where the intensity of convection is greater (Lawrence et al., 2007). Risks for human exposure in regions with NPPs are much more variable, largely determined by population densities, and

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- this level was about ten times higher than the ^{137}Cs deposition in Europe from global fallout;
- at this level the human dose during the first year after the core melt was about 1 mSv and was considered to be radiologically important.

5 Here we define $\geq 40 \text{ kBq m}^{-2}$ as “contaminated”, following IAEA (2005). Figure 2a shows the model calculated annual risks of contamination, from lower than 0.01 \% yr^{-1} in Alaska, the Sahara and Australia, to higher than 2 \% yr^{-1} in some areas around multiple reactors in the northeastern USA, western Europe and Japan. These numbers signify the expected values, defined as the weighted average (first moment) of an independent variable of all possible values it can take. The weights correspond to the probabilities of these values. This means, for example, that on average in the north-eastern USA, West Europe and Japan contamination by major accidents is expected at least every 50 yr. This is actually in accord with the frequency of past events in Europe and Japan. In Fig. 3 we present regionally enhanced sections of the same depiction as Fig. 2. There are extended regions with a risk of $> 1 \text{ \% yr}^{-1}$; and large parts of North America, East Asia and especially Europe have risks of more than 1 \% yr^{-1} , as indicated by the orange colour scale. In Fig. 2b and the lower panels of Fig. 3 we weigh the expected deposition with population density (SEDAC, 2011), which underscores the disproportional risk of contamination for people in regions with many reactors in Europe, parts of the USA and Asia.

20 Figures 4 and 5 show the results for ^{131}I , again defining $\geq 40 \text{ kBq m}^{-2}$ as “contaminated”. Interestingly, the deposition patterns over land and the risks of contamination and human exposure are quite comparable to ^{137}Cs . Even though the amount of radioactivity released by Chernobyl as ^{131}I was ~ 20 times that of ^{137}Cs , the low solubility of iodine and its ~ 1400 times shorter half-life limit deposition, especially on water surfaces. Recall that ^{131}I and ^{137}Cs are used as proxies for the total release and deposition of radionuclides, also including ^{90}Sr and ^{134}Cs , for example. By adding the risks of the two tracers in Figs. 2 and 4, the total risk of contamination by $\geq 40 \text{ kBq m}^{-2}$

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is roughly twice that indicated by the individual contamination risks. Furthermore, the risks scale with the addition of other tracers such as ^{134}Cs , ^{90}Sr , etc. However, also recall that we have taken the emissions by Chernobyl as a proxy for all 440 reactors. Preliminary estimates of the release of radioactivity during the first three weeks after the Fukushima accident are much lower than for Chernobyl, i.e. $\sim 140\text{ PBq}$ of ^{131}I and $\sim 10\text{ PBq}$ of ^{137}Cs (Chino et al., 2011). Stohl et al. (2011) estimated that during a period of 40 days after the accident $\sim 36\text{ PBq}$ of ^{137}Cs were released, about 42 % of the emission by Chernobyl. If the latter would be more representative of accidents associated with core melts than Chernobyl, the numbers in Figs. 2–5 would decrease by about a factor of seven. However, since these are only preliminary values, and do not include the full time period of release, it seems likely that these two estimates bound the actual probabilities.

It is important to underscore the different effects of ^{131}I and ^{137}Cs . In the initial period after accidents ^{131}I is of greatest concern as it deposits on agricultural crops, contaminating fruits, herbs and vegetables, and on grasslands where dairy cows graze (IAEA, 2006; Christodouleas et al., 2011). The contaminated grass ingested by the cows is transferred to the milk in about a day (Beresford et al., 2000). Even though the half-life of ^{131}I is only 8 days, it may elevate long term cancer risks (Christodouleas et al., 2011). According to Figs. 2–5 the integrated deposition loads of ^{131}I and ^{137}Cs are similar, while the latter exerts long term effects through its cycling and re-suspension in soils, groundwater and the vegetation. The half-life of ^{137}Cs (and ^{134}Cs and ^{90}Sr) is decades, and the main problem is the enduring exposure of the fauna through forage and humans through meat, milk and to a lesser extent vegetables (IAEA, 2006; WHO, 2006).

5 Footprint of catastrophic accidents

Next we calculated the footprint of nuclear accidents and find that the average surface area onto which $\geq 40\text{ kBq }^{137}\text{Cs}/\text{m}^2$ would be deposited in regions around a reactor

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after a core melt ranges from about 100 000 to 185 000 km². The smallest areas are in the Midwest-USA and the largest in South and East Asia. Much of the variability is related to the strength of vertical transport in convective storms and removal by precipitation, for example in the monsoon. This is consistent with the findings of Lawrence et al. (2007), who also showed that the venting of pollutants from the boundary layer is generally strongest in the tropics, whereas in the extra-tropics, where most NPPs are, the radionuclides will tend to remain more in the boundary layer over longer near-surface transport distances. Our model results indicate that the average number of people that would be affected by the radioactive contamination of ¹³⁷Cs due to a single reactor accident varies strongly by region: about 3 M in North Europe, 7 M in the Midwest USA, 11 M in East Europe, 14 M in East USA, 22 M in West Europe, 23 M in East Asia and 55 M in South Asia (M is mega = 10⁶).

For Chernobyl we computed a ¹³⁷Cs-contaminated area of about 113 000 km², endangering about 8 million people (note that this refers to 2005 mean meteorology and population statistics, not the 1986 simulation). This is not far from the published estimate for the Chernobyl accident, suggesting that an area of about 200 000 km² was contaminated, affecting more than 5 million people (Smith and Beresford, 2005), which provides additional support for the validity of our approach and its representativeness for other time periods. The model calculations suggest that some regions have a particularly high risk of contamination, due to the numerous reactors in operation, for example the region between Washington DC and New York, and around Atlanta, Toronto, Tokyo and Osaka. Many regions are densely populated and associated with a high human exposure risk, for example also Shanghai and Hong Kong. The highest risk of radioactive contamination occurs in West Europe, especially around the borders between Germany, Belgium and France, with the greatest human exposure risk in south-western Germany in the area between Stuttgart and Cologne.

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6 Phase-out of NPPs in Germany

In the wake of the events in Fukushima the German government has decided to phase out all nuclear power plants over the next decade (in accord with the theory that large-impact and rare events are leading causes of societal change; see Taleb, 2010). Figure 6a presents calculations in which the 17 German reactors have been switched off. This reduces the expected ¹³⁷Cs deposition in Germany by about a factor of two, though it nevertheless continues to be among the highest values worldwide. Only in France (58 reactors) and Belgium (7 reactors) does the overall risk remain higher. Figure 6b shows that by switching off the reactors in neighbouring countries, i.e. France, Belgium, the Netherlands, Switzerland and the Czech Republic, but leaving all 17 German reactors active, the deposition risk in Germany would be even more effectively reduced than by the national efforts alone. This emphasizes that, for the sake of reducing the risk of exposure to radioactivity, there is a need to coordinate phase-out decisions on an international level, especially when NPPs are located in the vicinity of international borders.

7 Conclusions and recommendations

Using our approach, it will be possible to evaluate other risks besides direct human exposure, including biological and indirect health risks. For instance, linking to agricultural information will allow evaluation of the risk that radioactivity enters the food chain. Clearly there is a need to perform an extensive re-assessment of the factors (a) and (b), discussed above, based on the evidence from Chernobyl, Fukushima and other less catastrophic accidents. In particular, a better understanding of reactor risk profiles and expected release of radioactivity in case of a meltdown is required. This depends on several aspects, including:

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- type of reactor and capacity;
- reactor maintenance and other human factors;
- safety improvements with the progress of technology;
- degradation of the reactor shell with age;
- enhanced risk for plants with multiple reactors and shared technical facilities;
- the likelihood of natural disasters such as earthquakes and tsunamis;
- susceptibility to aircraft impacts, sabotage and terrorist attacks.

Key information in the assessment of exposure risks is the emission strength of radioactivity by major nuclear accidents. We have used the available reports of the Chernobyl disaster, and it will be important to also evaluate the Fukushima accident at the same level of detail. Preliminary estimates suggest that the Fukushima emissions of radioactivity are up to an order of magnitude less than of Chernobyl. If representative for NPPs, this would reduce the risks presented here correspondingly and the present risk assessment should be considered as a worst case scenario. However, since most of the radioactivity from Fukushima has been deposited to the Pacific Ocean, remote from measurement stations, the inverse computation of emissions based on observational data will be difficult, although the back-calculation of trans-Pacific transport based on measurements along the US west coast holds promise (Priyadarshi et al., 2011). Furthermore, it will be important to substantiate the level of “dangerous” contamination. We primarily applied 40 kBq m^{-2} of ^{137}Cs , but this does not do justice to the acute dangers by the short-lived ^{131}I and the additional contamination by other long-lived radionuclides.

Going beyond the factors (a) and (b), we have provided a better understanding of the impact of atmospheric dispersion, providing evidence that the widespread risks to humanity of nuclear accidents are much larger than suggested in official reports

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several decades ago. It will be important to scrutinize reactor sites by accounting for the proximity to large population centres, as there is a tendency to build NPPs in the vicinity of electricity consumers in urban regions. NPPs in densely populated areas in Europe, the USA, eastern Asia and especially southern Asia bear a high risk of exposing large numbers of people to radioactive contamination after reactor accidents. In western Europe (notably France), the concentration of NPPs is relatively high, and although the phase out in Germany in the next decade will halve the national risk of radioactive contamination by major accidents, international efforts will be necessary to fully achieve the German safety objectives. Worldwide more than 60 reactors are currently under construction (Supplement) and many more are planned, hence the global risks may change accordingly in the coming years.

Supplementary material related to this article is available online at:
**[http://www.atmos-chem-phys-discuss.net/11/31207/2011/
acpd-11-31207-2011-supplement.pdf](http://www.atmos-chem-phys-discuss.net/11/31207/2011/acpd-11-31207-2011-supplement.pdf)**

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**Table 1.** Radioactivity released to the atmosphere by INES 4–7 nuclear accidents (in PBq).

Location	Country	INES	Date	Total	¹³¹ I	¹³⁷ Cs
Fukushima	Japan	7	11 Mar 2011	> 630	> 150	12–35.8
Chernobyl	USSR	7	26 Apr 1986	> 12 000	1760	85
Mayak	USSR	6	29 Sep 1957	74–1850	n.d.a	n.d.a
Chalk River	Canada	5	12 Dec 1952	> 0.1	n.d.a.	n.d.a.
Windscale	UK	5	10 Oct 1957	1.6	0.7	0.02
Simi Valley	USA	5	26 Jul 1959	^b	^a	n.d.a.
Lucens	Switzerland	4–5	21 Jan 1969	^d	^d	^d
Belojarsk	USSR	5	1977	n.d.a.	n.d.a.	n.d.a.
Three Mile Island	USA	5	28 Mar 1999	1.6*	< 0.0007	n.d.a.
Chernobyl	USSR	5	1 Sep 1982	n.d.a.	n.d.a.	n.d.a.
Idaho Falls	USA	4	29 Nov 1955	^d	^d	^d
Idaho Falls	USA	4	3 Jan 1961	n.d.a.	n.d.a.	n.d.a.
Monroe	USA	4	5 Oct 1966	^d	^d	^d
Windscale	UK	4	1973	n.d.a.	n.d.a.	n.d.a.
Leningrad	USSR	4	6 Feb 1974	^e	n.d.a.	n.d.a.
Leningrad	USSR	4	Oct 1975	55	n.d.a.	n.d.a.
Jaslovské Bohunice	CSSR	4	22 Feb 1977	n.d.a.	n.d.a.	n.d.a.
Saint-Laurent	France	4	13 Mar 1980	n.d.a.	n.d.a.	n.d.a.
Buenos Aires	Argentina	4	23 Sep 1983	^c	^c	^c

n.d.a. no data available,

^a data for ¹³³Xe available,^b main emission ⁸⁵Kr, ¹³³Xe,^c strong ¹³¹I emission assumed, though n.d.a.,^d no radioactivity released to the atmosphere,^e release of radioactive sludge from filter powder to the environment,* mainly ⁸⁵Kr emitted.

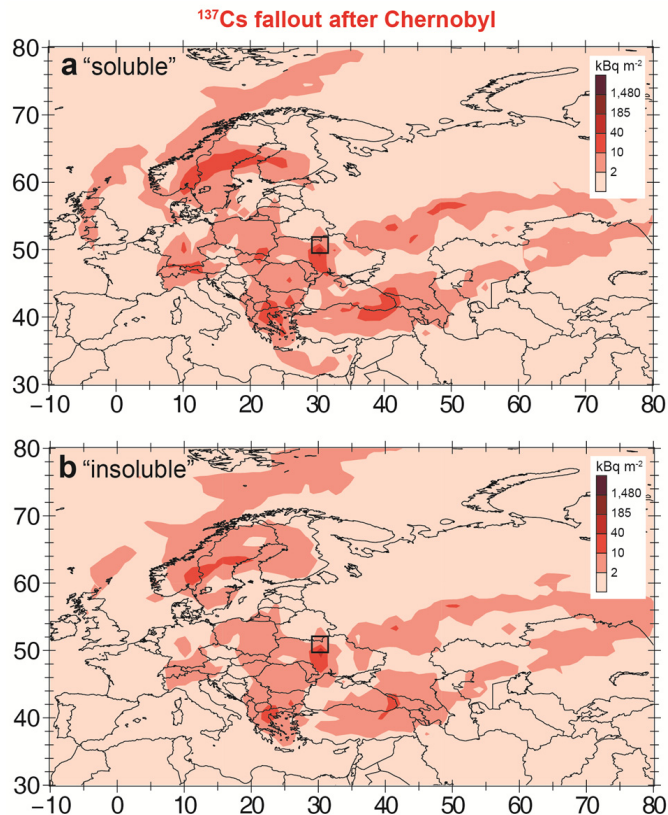


Fig. 1. Model calculated fallout of ^{137}Cs after the Chernobyl accident, assuming a soluble particle tracer **(a)** and an insoluble one **(b)**. The location of Chernobyl is indicated by the black square. The colour scale is the same as in Peplow (2006).

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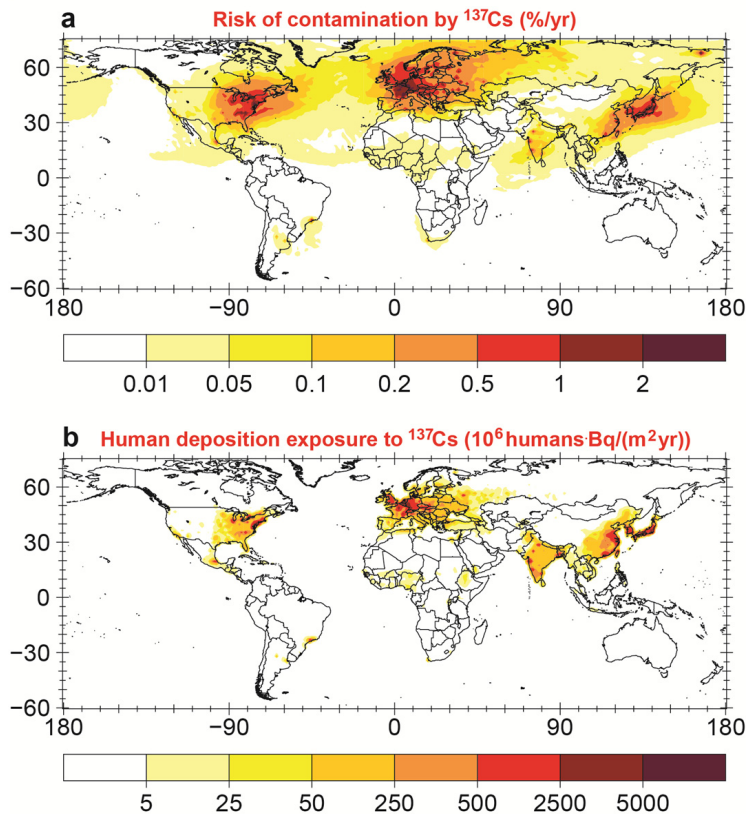


Fig. 2. Global risk of radioactive contamination by ^{137}Cs . **(a)** Modelled deposition of $\geq 40 \text{ kBq } ^{137}\text{Cs}/\text{m}^2 \text{ yr}^{-1}$. The risk is the expected value normalized by 40 kBq m^{-2} . **(b)** Modelled risk of human exposure to ^{137}Cs deposition.

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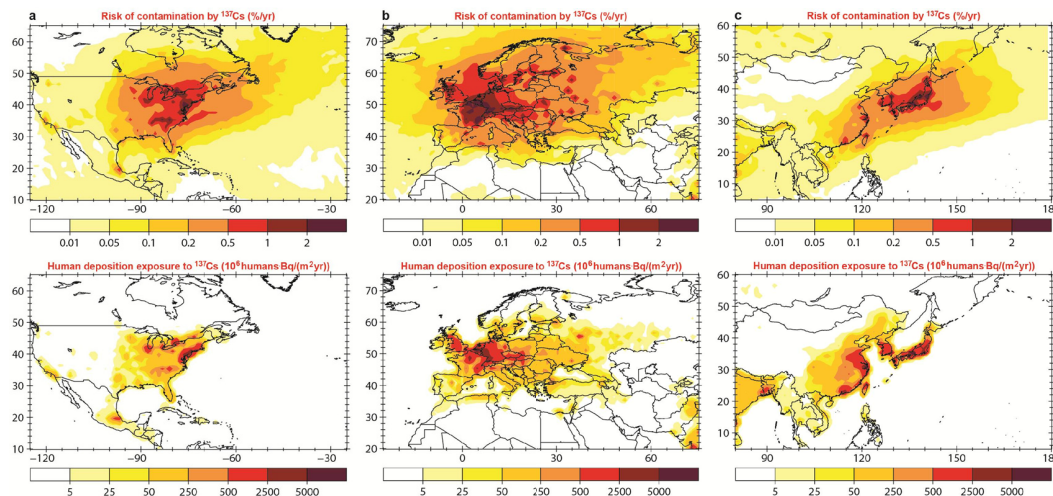


Fig. 3. Regional risks of radioactive contamination by ^{137}Cs . Same as Fig. 2 for selected regions.

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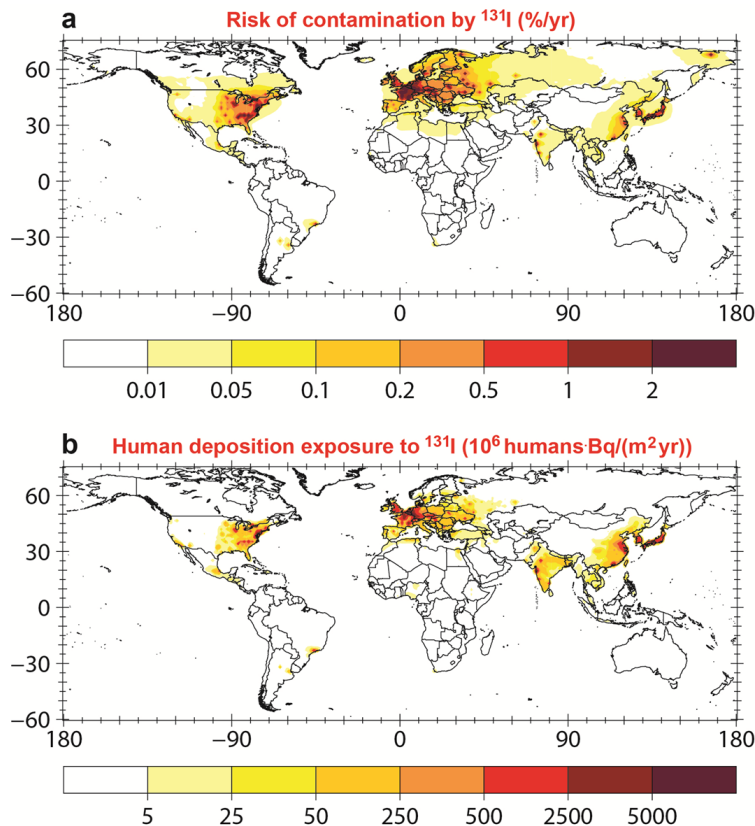


Fig. 4. Global risk of radioactive contamination by ^{131}I . **(a)** Modelled deposition of $\geq 40 \text{ kBq } ^{131}\text{I}/\text{m}^2 \text{ yr}^{-1}$. The risk is the expected value normalized by 40 kBq m^{-2} . **(b)** Modelled risk of human exposure to ^{131}I deposition.

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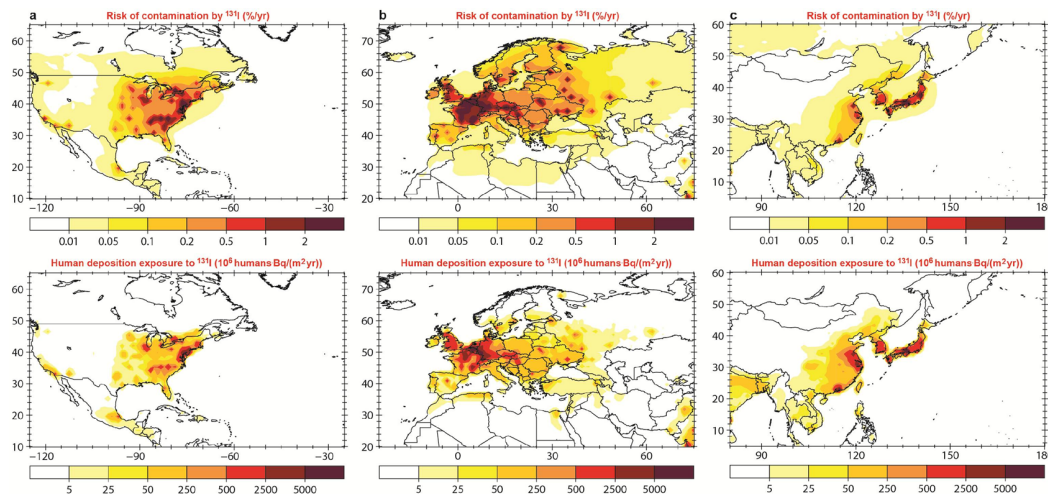


Fig. 5. Regional risks of radioactive contamination by ^{131}I . Same as Fig. 4 for selected regions.

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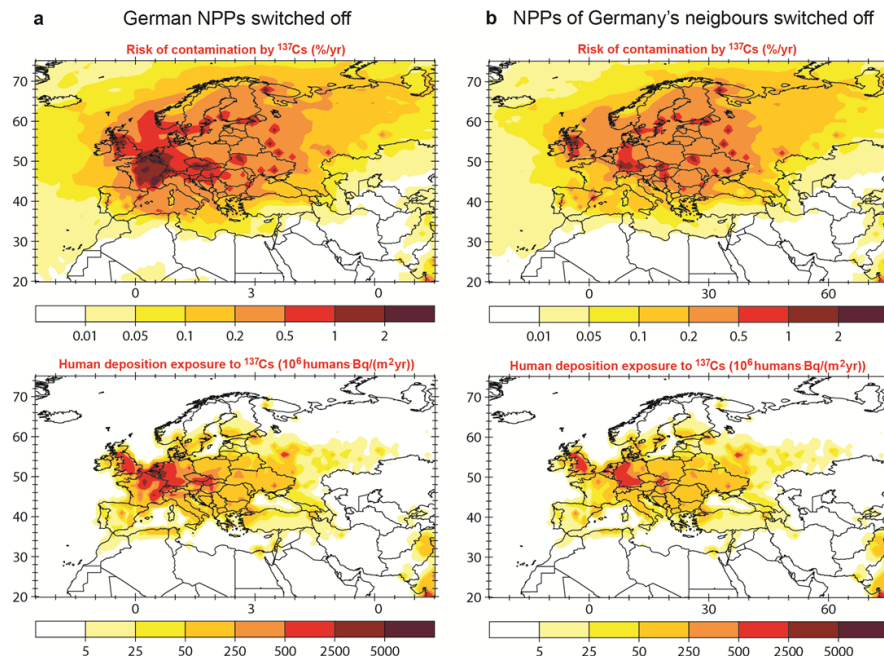


Fig. 6. Phasing out nuclear power plants. **(a)** Same as Fig. 3b, indicating the regional risks of contamination by ^{137}Cs with German reactors switched off. **(b)** Same as Fig. 3b with reactors in the neighbouring countries of Germany switched off.