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[C. Schnabel](#), [J. M. López-Gutiérrez](#), [Sönke Szidat](#), [M. Sprenger](#) ...+3 more authors

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On the origin of ^{129}I in rain water near Zürich

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I-129 / Sellafield / La Hague / ^{129}I / Atmospheric transport

anthropogenic emissions primarily from nuclear fuel reprocessing plants, which are discussed in detail below. A review of modern $^{129}\text{I}/^{127}\text{I}$ ratios in the environment is given by Schmidt *et al.* [2]. Here, only the nearly continuous increase of ^{129}I concentrations in an ice core from the Swiss Alps from 1950 to 1986 is mentioned [3, 4]. A calculated mean $^{129}\text{I}/^{127}\text{I}$ ratio for the years 1983 to 1986 for that ice core is as high as 5×10^{-8} assuming an iodine content of $1.75 \mu\text{g}/\text{kg}$, a mean concentration calculated for precipitation in southern Germany (see below). When these data from a high altitude sampling site are compared to data from Bachhuber and Bunzl [5] for rain in Bavaria/Germany 1988/89 with $^{129}\text{I}/^{127}\text{I}$ ratios from 6.2×10^{-7} to 9.6×10^{-7} , a further dramatic increase can be seen. Then, the environmental $^{129}\text{I}/^{127}\text{I}$ ratios in central Europe seem to have remained stable in the early 1990s [6, 7]. In spite of the fact that all these ^{129}I abundances far away from the emission sources are not seen as radiologically hazardous to people, a detailed observation of the trends of ^{129}I concentrations in Europe is considered necessary. This necessity has become even more important due to the fact that one of the main sources of ^{129}I releases into the environment in the 1980s – the nuclear fuel reprocessing facility in La Hague – has increased its liquid discharges of ^{129}I into the English Channel drastically after 1988 [8] whereas the liquid releases from Sellafield were increased much less from 1988 to 1996 [9, 10]. Moreover, as shown in detail below, both reprocessing plants continue with their ^{129}I discharges at a very high level.

The equilibrium mass of naturally produced mobile ^{129}I in the compartments atmosphere, biosphere, and hydrosphere was estimated at 260 kg [11]. This reservoir was enlarged by atmospheric nuclear weapons tests. For these releases, ^{129}I masses of 50 kg [12], 80 kg ([13, 14]) or 150 kg ([13, 15]) are published. Eisenbud and Gesell [13] give 1.9 GBq ^{129}I emissions per megaton atmospheric bombs. For the yields of atmospheric bomb tests, the estimates vary from 276 megatons [14] to 547 megatons [15]. The Chernobyl accident gave only rise to an atmospheric emission of 1.3 kg ^{129}I [16]. In contrast, Hanford (nuclear fuel reprocessing plant in the north-western USA) emitted from 1944 to 1972 around 260 kg ^{129}I into the atmosphere [17] with high peak emissions in the mid 1940s to early 1950s,

* Author for correspondence
(E-mail: schnabel@particle.phys.ethz.ch).

† Present address: Depto. de Física Aplicada I. Escuela Universitaria Politécnica, Universidad de Sevilla, Spain.

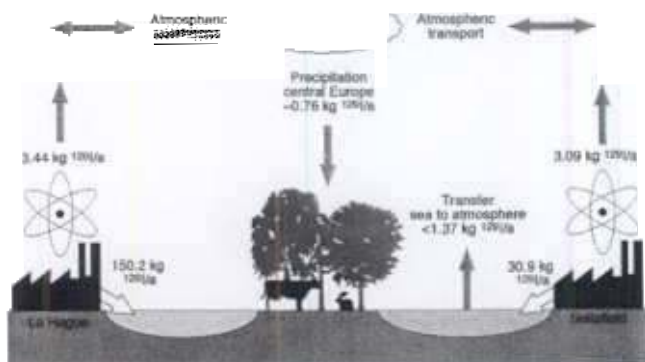


Fig. 1. Schematic illustration of some ^{129}I emission and immission data as annual averages from 1991 to 1996.

3. Results and discussion

The radionuclide concentrations of ^{129}I determined in the rain water samples are shown in Table 1.

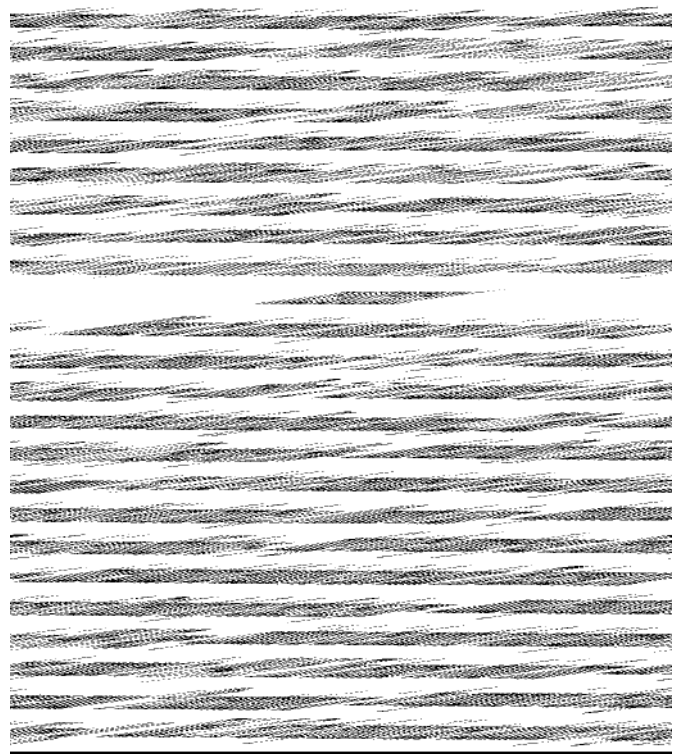
A large scatter in monthly data is observed. Similar variations have also been measured by Krupp and Aumann for rainfall in Germany [22] and at a shorter time resolution by Lopez-Gutierrez *et al.* for rain in southern Spain [24]. A possible reason for this scatter will be discussed in connection with the application of a meteorological transport model below. The mass-weighted annual mean concentrations of ^{129}I are compared to literature data for central Europe in the 1980s and 1990s in Table 2.

Mass-weighted average relative standard deviations (one sigma) for the annual ^{129}I concentrations at Dübendorf range from 3.7% to 4.3%. Because the original ^{127}I concentration of the rain was not analyzed, an estimate of the inherent $^{129}\text{I}/^{127}\text{I}$ ratio of the samples should be based on an assumption of the ^{127}I concentration. The compilation of Fuge and Johnson [25] gives a range from 0.5 to 10 $\mu\text{g}/\text{kg}$ I in rain, whereas from the data from Krupp and Aumann [22] an average concentration of 2 $\mu\text{g}/\text{kg}$ can be calculated for southern Germany, compared to about 1.5 $\mu\text{g}/\text{kg}$ for a site in Bavaria calculated from the data of Bachhuber and Bunzl [5]. Consequently we choose 1.75 $\mu\text{g}/\text{kg}$ I as basis to estimate the inherent $^{129}\text{I}/^{127}\text{I}$ ratio of the samples analyzed. Moran *et al.* [26] determined 1.7 $\mu\text{g}/\text{kg}$ as median iodine concentration of precipitation samples in the US. The mass-weighted means for the inherent $^{129}\text{I}/^{127}\text{I}$ ratios are: 3.8×10^{-7} for 1994, 3.2×10^{-7} for 1995 and 4.0×10^{-7} for

Table 1. ¹²⁹I atom concentrations in water samples from Dübendorf/Zürich, Switzerland.

Date	Concentration (atoms/l)	Date	Concentration (atoms/l)
		June 1995	26.1 ± 1.3
		July 1995	16.3 ± 0.7
		August 1995	26.9 ± 1.1
		September 1995	10.1 ± 0.4
		August 1996	5.35 ± 0.3
		September 1996	14.9 ± 0.5
		October 1996	52.0 ± 2.0
		November 1996	39.8 ± 0.8
		December 1996	7.45 ± 0.20
		January 1997	70.4 ± 3.0
		February 1997	25.4 ± 2.7
		March 1997	92.8 ± 4.4
		May 1997	445 ± 13
		June 1997	22.2 ± 0.4

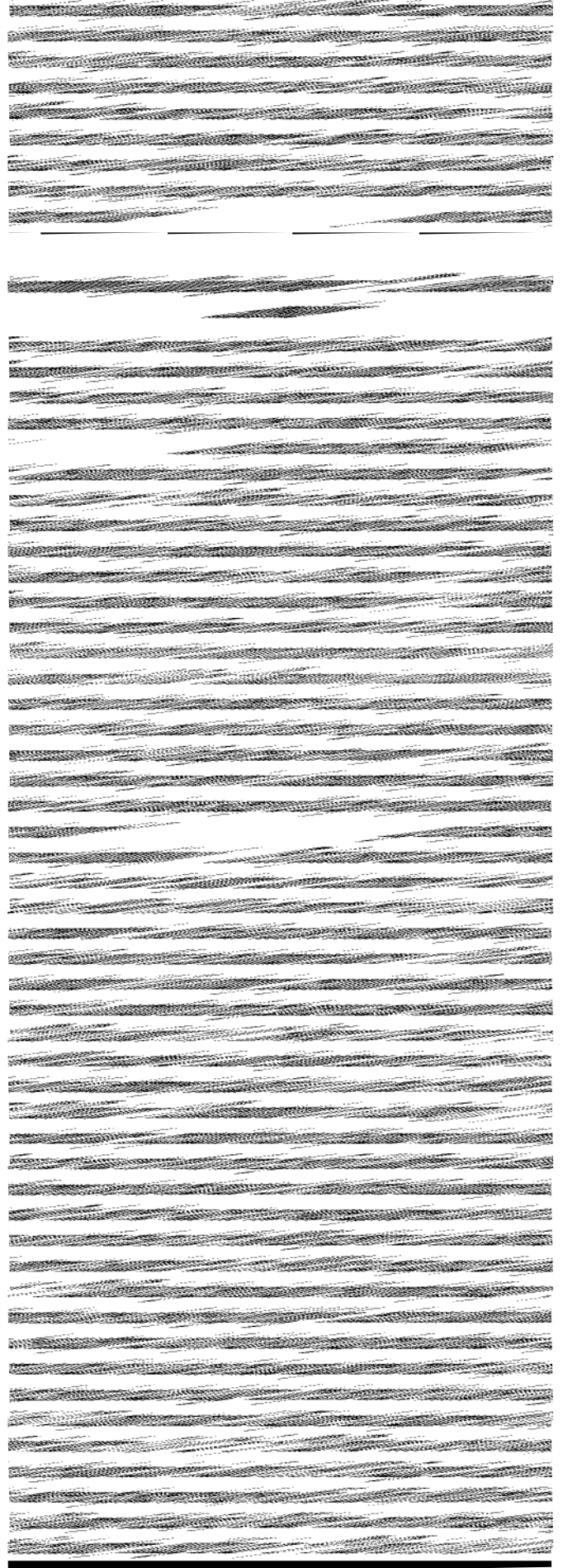
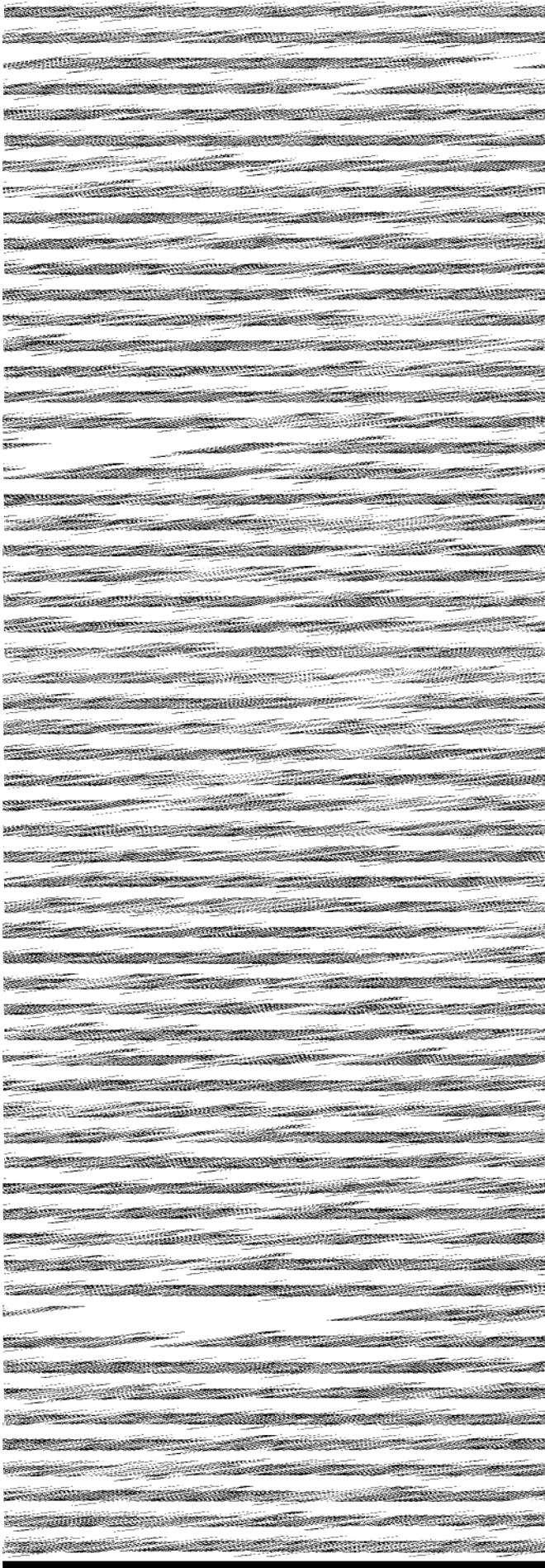
Reference	Concentration (atoms/l)
[1]	3.30–4.66
[2]	12.1–62.6
[3]	37.5
[4]	76.6
[5]	31.6
[6]	26.3
[7]	33.1
[8]	60.7–88.7



1996/97, with relative standard deviations of about 4% for each ratio.

These annual mean isotopic ratios are of the same order of magnitude but slightly lower than those determined by

Location	Year	Concentration (atoms/l)	Reference
	1983–86	0.6–0.9	[9]
	1988	4.0	[5]
	1989	6.6	[5]
	1995	5.8–10.6	[22]
	1994	5.34	This work
			This work
		2.31	This work
	1997		
	1998	3.9–6.6	
	1999 (Jan–Jun)	1.9–3.4 (calculated from first 6 months)	



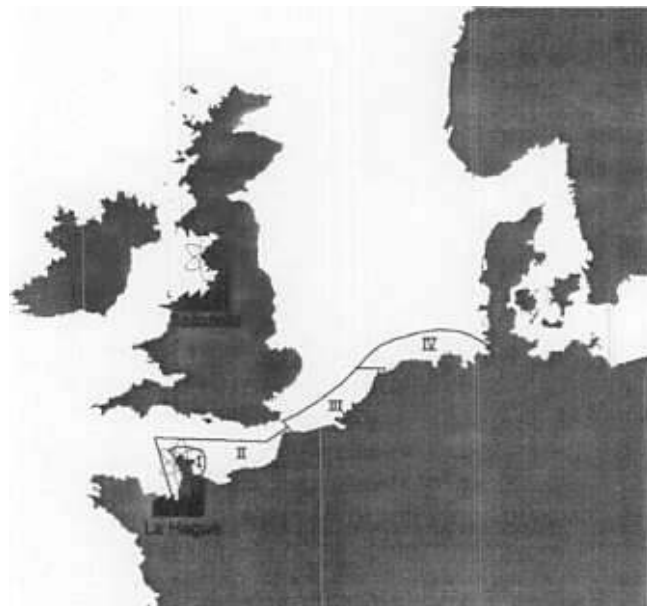
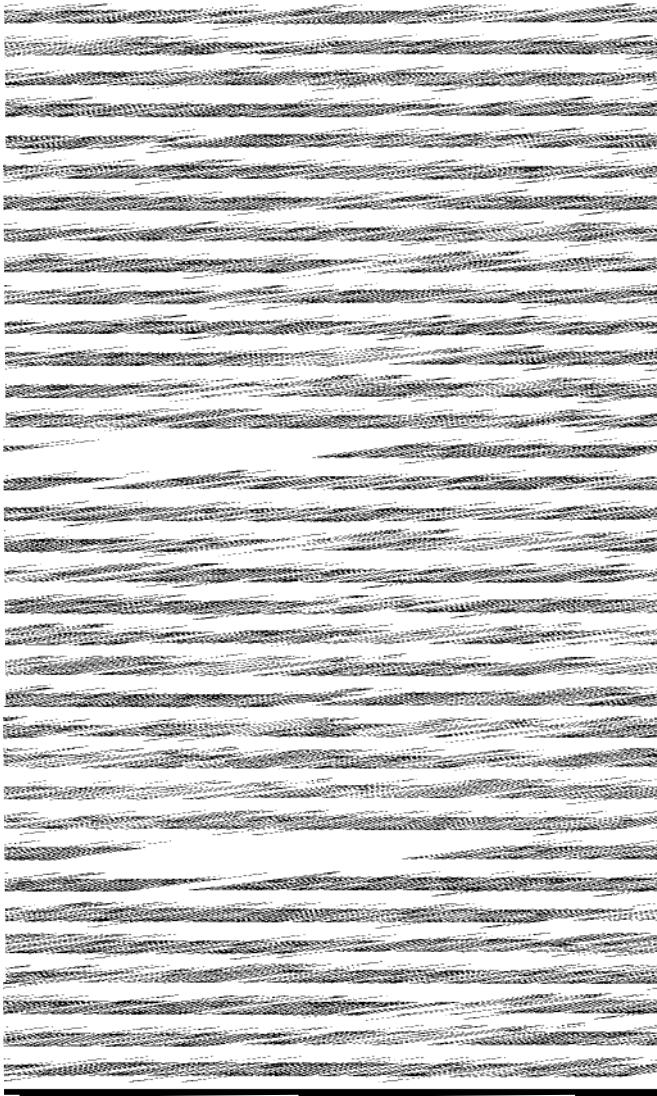
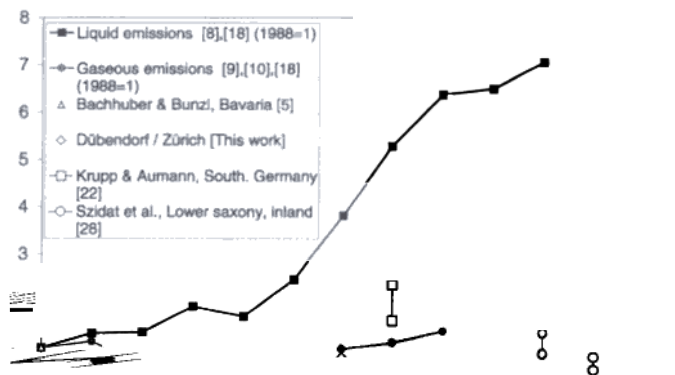


Fig. 2. Areas of the English Channel and coastal areas of the North Sea used as basis to estimate the area with an $^{129}\text{I}/^{127}\text{I}$ ratio of around 5×10^{-7} at the early 1990s. The $^{129}\text{I}/^{127}\text{I}$ ratios taken for the different zones are: 2.5×10^{-6} for zone I, 5×10^{-7} for II, 3.1×10^{-7} for III, and 1.6×10^{-7} for IV.

Europe. For the iodine flux from the sea to the atmosphere, the publication of Rasmussen *et al.* [32] is used as basis. These authors give a flux of $1 \times 10^{+9}$ kg I/a from areas of high biological activity that amount to only 10% of the global sea surface, *i.e.* 3.5×10^7 km². Using that flux density of 28.6 kg I/km², the affected sea surface of 96 000 km² calculated above and an isotopic ratio of 5×10^{-7} , 1.37 kg ^{129}I are estimated as an upper limit for the mass annually transferred from the affected sea area. We would like to emphasize that we treated the whole area in Fig. 2 as well as a region in the Irish Sea as regions of high biological activity. For the precipitation in central Europe about 760 g ^{129}I per year are needed, when an area of 10^6 km² and an annual deposition rate of $5 \text{ mBq m}^{-2} \text{ a}^{-1}$ are used as basis. For a more reliable estimate, the iodine transfer in the North Sea and the English Channel itself has to be studied. Nevertheless, about 6.5 kg ^{129}I [9, 10, 18] emitted annually from Sellafield and La Hague to the atmosphere from 1991 to 1996 (without 1993 for which we have no data for Sellafield) can preliminarily be compared to this 1.37 kg. Based on this comparison, it is very likely that the gaseous emissions do contribute substantially to the precipitation in central Europe even when the large uncertainties of the comparison are taken into account. It is emphasized again that a more precise determination of the transfer factor for iodine from the sea to the atmosphere at the location of interest itself is needed to better estimate the relative contributions of liquid and gaseous discharges to the precipitation in central Europe.

To answer the third question about the correlation of the time dependences of gaseous and liquid ^{129}I emissions with the ^{129}I concentrations in precipitation in central Europe since 1988 the respective data are plotted in Fig. 3. In that figure, ^{129}I concentrations in precipitation as well as both types of releases are normalized to their respective values in 1988. The liquid discharges were enhanced by a factor of 6.4 from 1988 to 1996. This increase has been observed, as stated above, in seaweed samples at the coast of south-



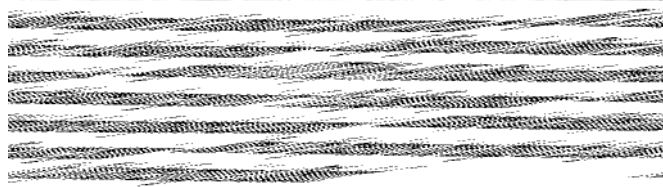
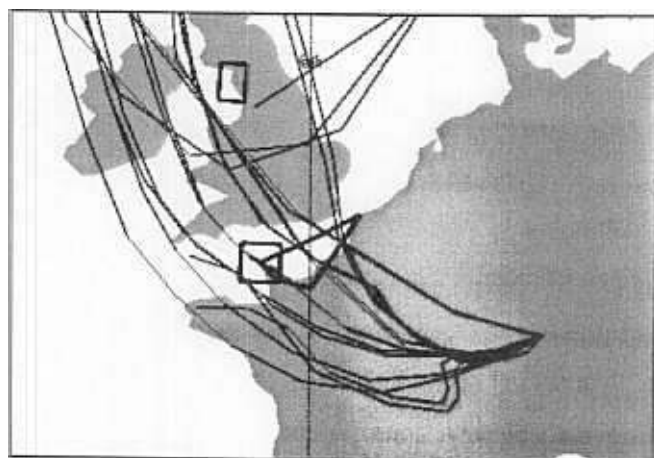
ern Norway [8]. In contrast to this the gaseous emissions from Sellafield and La Hague remained approximately constant during that period and exhibited only an increase by a factor of 1.4 from 1988 to 1996 [9, 10, 18]. The ^{129}I concentrations determined in central Europe since 1988 show only a weak, if any, time dependence. Consequently, the ^{129}I concentrations in precipitation are in much better agreement with the time dependence of the gaseous emissions of the two reprocessing plants than with their liquid releases.

The month-to-month variations in ^{129}I concentrations (cf. Table 1) might be related to differences in the source regions and atmospheric transport conditions of the air masses that lead to the sampled precipitation close to Zürich. For instance, large monthly ^{129}I concentrations might be a consequence of many rain events during the months with advection directly from La Hague or from Sellafield and with no or only a few rain events with transport from a different source region which would dilute the measured ^{129}I precipitation concentrations. Additionally for the air masses advected from the La Hague/Sellafield region it is important that no (or little) precipitation occurs before they reach the measurement site. On the other hand, low monthly ^{129}I concentrations can be expected when the air mass advection is not from the La Hague/Sellafield region on rainy days in Zürich, or when the air parcels have lost their ^{129}I atoms due to wet scavenging on their way to central Europe, or when significant dilution occurs from rain events with other source regions.

In order to test the influence of atmospheric transport, three-dimensional assimilated wind fields from the European Center for Medium-Range Weather Forecast (ECMWF) are used to calculate backward trajectories [34] for two high ^{129}I months May 1994 (9405) and May 1997 (9705), and two low ^{129}I months November 1994 (9411) and December 1996 (9612). The wind fields are globally available every 6 hours on a latitude/longitude grid with a horizontal resolution of 1 degree and on 31 vertical levels (therefrom about 14 in the lowest 5 km). Note that assimilated data constitute a "sophisticated combination" of a short-range numerical model forecast and all globally available surface and upper-air observations. A set of 19 trajectories is started between 950 hPa and 400 hPa at Dübendorf every 6 hours during a selected month, and then calculated 72 hours backward in time. Daily precipitation data for Dübendorf was provided by the ANETZ measurement station of the Swiss Meteorological Institute in Zürich, which is situated about 10 km from Dübendorf. The total amount of precipitation for the selected four months are: 234.2 mm in 9405, 54.3 mm in 9705, 43.0 mm in 9411, and 78.5 mm in 9612.

Fig. 4 shows an example for atmospheric transport from the La Hague/Sellafield region to the measurement site on 8 May 1997. Furthermore, two regions around each of the source regions La Hague and Sellafield are marked, which we considered for liquid and gaseous emissions.

The results of the trajectory calculations are summarized in Table 4a and 4b, which are based upon those trajectories that satisfy the following two criteria: The backward trajectory started at Dübendorf hits at least one of the four source regions marked in Fig. 4, and the precipitation in Zürich exceeds 1 mm on the starting day of the trajectory. Table 4a



	9405	9411	9612	9705
La Hague marine	6	2	3	1
La Hague atmospheric	6	3	2	1
Sellafield marine	1	2	1	1
Sellafield atmospheric	1	2	1	1

Table 4b. The percentage of monthly precipitation in Zürich which is associated with condensation in air masses coming from one of the four source regions. The analysis is based upon the same trajectories as the ones used in Table 4a.

	9405	9411	9612	9705
La Hague marine	22	27	19	33
La Hague atmospheric	23	33	15	19
Sellafield marine	2	15	7	10
Sellafield atmospheric	2	30	7	10

gives the number of days per month where these two criteria are fulfilled. During the high ^{129}I months May 1994 and May 1997, the La Hague source region is more often "connected" to the measurement site than during the low ^{129}I months Nov. 1994 and Dec. 1996. On the other hand, this is not true for Sellafield, for where no significant differences can be discerned. Further information can be gained from Table 4b which answers the question how much of the monthly precipitation can be potentially loaded with ^{129}I . For instance, 22% of the precipitation which was measured in May 1994 in Zürich, was due to condensation in air masses coming from the La Hague marine region. If differences in the source regions and atmospheric transport conditions

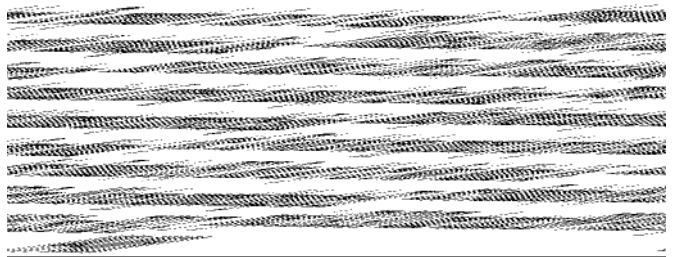
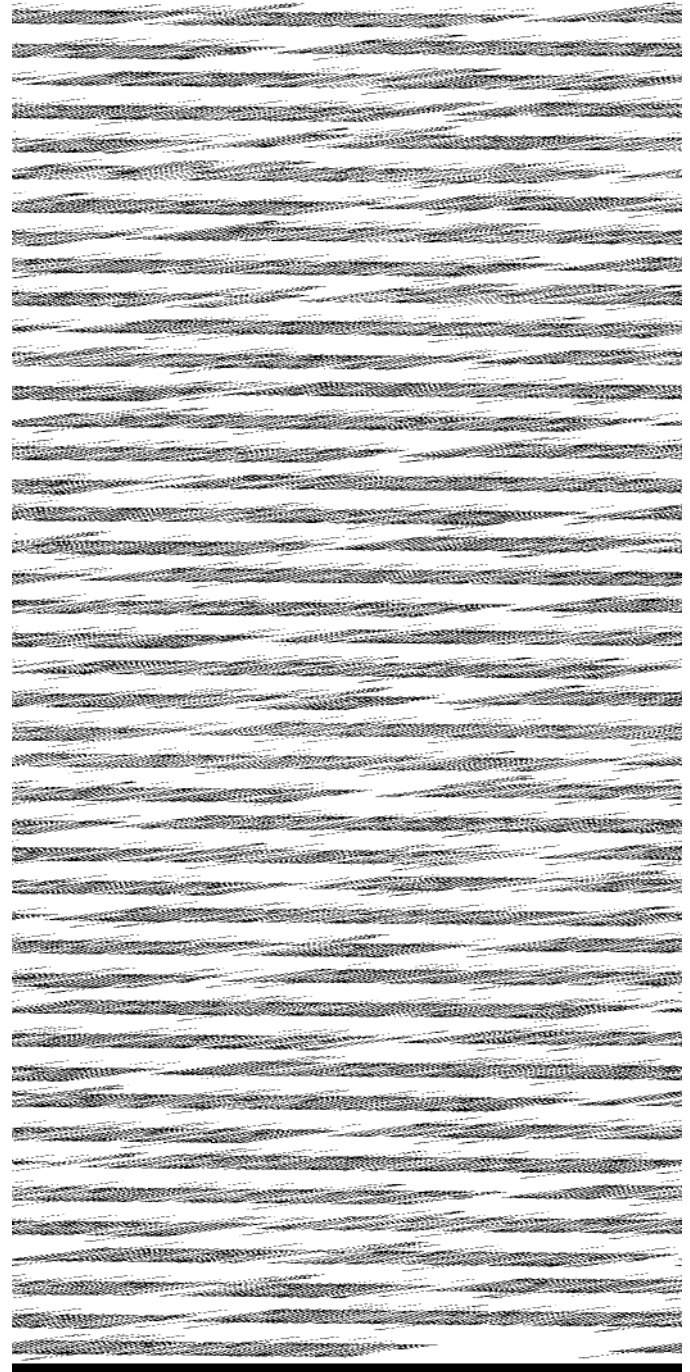
alone were able to explain the large month-to-month variations, we would expect to see similar variations in Table 4b as for ^{129}I concentrations in Table 1. However, the variability which can be attributed to atmospheric transport is much smaller than what has been observed for ^{129}I concentrations in precipitation, when constancy of the gaseous emissions with time is assumed.

Two possible explanations for this discrepancy relate to the atmospheric transport from the source regions to the measurement site itself, and to the variability of the gaseous release of ^{129}I . These two issues will be briefly discussed. We assumed that the emitted ^{129}I (for instance from La Hague) will be rained out in Zürich if the trajectory "connects" the source with the measurement site. But wet scavenging of an air parcel before its arrival at the measurement site might, in fact, considerably reduce the amount of ^{129}I rained out later in Zürich. The consideration of this effect is, however, very difficult given the coarse temporal resolution (24 hours) of the precipitation measurements over Europe. For the second issue, additional qualitative information can be gained from weekly ^{85}Kr measurements in Freiburg im Breisgau [35], which is located about 120 km from Zürich. High ^{85}Kr concentrations may indicate high gaseous ^{129}I emissions, whereas low ^{85}Kr concentrations point to low gaseous ^{129}I discharges. It must however be emphasized that ^{85}Kr unlike ^{129}I is not washed out by precipitation, and that the distance between Freiburg and Zürich allows only qualitative inferences. In the high ^{129}I months, precipitation associated with transport from La Hague/Sellafield occurs during the weeks with high ^{85}Kr concentrations (for instance, the precipitation event on 22 May 1997 goes along with a weekly ^{85}Kr value of 2.73 mBq/m^3 , and the one on 28/29 May 1994 with 1.97 mBq/m^3). On the other hand, lower ^{85}Kr concentrations (mostly below 1.4 mBq/m^3) are typical for the low ^{129}I months.

To summarize, the trajectory-based analysis cannot account for the large month-to-month variations of the ^{129}I of two orders of magnitude, and suggests that they could be due to variable gaseous emissions or due to wet scavenging between the source regions and Zurich. More detailed investigations would be highly desirable, if possible based upon weekly ^{129}I samples and upon the now published monthly data of gaseous halogen emissions [36], that are available on the internet. These new release data for La Hague indicate that the gaseous ^{129}I discharges have been decreased by about a factor of 6 from 1996 to 2000. Unless a similar reduction was achieved at Sellafield, the British re-processing plant should dominate the gaseous ^{129}I releases in 2000.

A last attempt to distinguish between marine or atmospheric ^{129}I is to look at the seasonal variation in our data. Whereas four of the five highest monthly ^{129}I concentrations were found in spring, Rasmussen *et al.* [32] found a maximum in methyl iodide transfer from the sea to the atmosphere during summer. Consequently, we tend to assume that high ^{129}I concentrations in spring do not reflect an increase in biological activity in the North Sea, but may be more likely due to high gaseous discharges during the respective months as indicated by ^{85}Kr measurements in Freiburg im Breisgau [35] and by monthly ^{85}Kr emission data from La Hague [36].

4. Conclusions



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