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Distribution of  $^{127}\text{I}$  and  $^{129}\text{I}$  in precipitation at high European latitudes

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We here present the most extensive data set on the distribution of  $^{127}\text{I}$  and  $^{129}\text{I}$  in precipitation (rain and snow) covering the period 2000-2006 and European latitudes  $55^{\circ}\text{N}$ - $68^{\circ}\text{N}$ . Our results indicate a wide variation in the concentrations and fluxes of the two isotopes associated with generally higher values at near coastal sites compared to the inland ones. Total wet-related annual deposition of  $^{127}\text{I}$  and  $^{129}\text{I}$  on Sweden and Denmark is estimated at about  $1.2 \times 10^9$  g and 60 g respectively. The average annual  $^{129}\text{I}$  wet deposition accounts for  $< 1\%$  and  $< 0.05\%$  of the total annual gaseous and liquid, respectively, discharges from the Sellafiled and La Hague Facilities. The  $^{127}\text{I}$  annual wet deposition represents  $< 1\%$  of the estimated global oceanic iodine flux. Air mass trajectories suggest that events of enhanced  $^{129}\text{I}$  in precipitation are closely related to southwesterly weather fronts from regions of elevated concentrations.

## **1. Introduction**

Anthropogenic production of  $^{129}\text{I}$  from  $^{235}\text{U}$  nuclear and  $^{239}\text{Pu}$  nuclear fission, common components of spent nuclear fuel, is a major source of the isotope in the earth's surface environments. About 5500 kg of  $^{129}\text{I}$  have been released since the beginning of the atomic era in early 1940s and tremendous amounts (estimated at 70000 kg) of  $^{129}\text{I}$  still pending

in unprocessed nuclear fuel with new nuclear reprocessing facilities planned in future. Although iodine is an essential element for human health, the current high level of the  $^{129}\text{I}$  in the environment means that understanding of major fluxes and pathways becomes indispensable for planning of future environmental monitoring and mitigation efforts. Oceans are the major reservoir and source of stable iodine and the question of whether coastal regions may eventually receive more iodine deposition than inland region remains uncertain. It has been suggested that emission from algal growth, and in particular macro algae along coastal region, is a reason behind the enrichment (up many thousands times) of the marine boundary layer in iodine compared to the original seawater (O'Dowd et al., 2002; McFiggans et al., 2004). On land iodine emission from rice paddies and wetlands is considered most important (Redeker et al., 2000), but recent studies point out large iodine emissions from middle latitude vegetation and soils (Sive et al., 2007).

Several investigations point out iodine emissions as a significant contributor to aerosol budget, and in particular the marine aerosol, which can form micro particles that become cloud condensation nuclei (Baker et al., 2001; Pirjola et al., 2005; Saiz-Lopez et al., 2006). With sufficient influence on the extent of marine aerosol formation, the huge amounts of iodine marine (bio-) emissions ( $10^{12} \text{ g y}^{-1}$  or  $1.4 \times 10^3 \text{ atoms cm}^{-2} \text{ s}^{-1}$ ) are suggested to have direct affect on the global climate forcing. Furthermore, it is presently acceptable that the overwhelming anthropogenic supply, discharge type (liquid and gases) and chemical forms (dominantly iodide, iodate or  $\text{I}_2$ ) of  $^{129}\text{I}$  create some doubts about the isotopic equilibrium and chemical association of  $^{129}\text{I}$  and  $^{127}\text{I}$  in the environment (Hou et al. 2007).

The atmosphere is one of the most sensitive environmental compartments for natural and anthropogenic changes and provides rather rapid response to disequilibrium in the system. Sources of the stable iodine to the atmosphere are still mainly of natural origin on global scale (knowing that extensive rice paddies and a future use of methyl iodide as soil fumigant adds an anthropogenic component). Conversely sources and impact of anthropogenic  $^{129}\text{I}$  on the atmosphere may globally be rather different from that of  $^{127}\text{I}$  (Snyder and Fehn, 2004; Aldahan et al. 2007). Generally, the atmosphere of the northern hemisphere contains about 10-100 folds more  $^{129}\text{I}$  than the southern one, and also there is a large variability within the northern hemisphere regions. Several studies (Aldahan et al. 2007 and references therein) suggest up to 1000 times higher concentrations in the European atmosphere compared to that of North America. The main reason behind this global variability of  $^{129}\text{I}$  is related to the huge liquid and gaseous releases from the nuclear fuel reprocessing facilities and in particular the ones located at the western European coasts, namely, at La Hague (France) and Sellafield (UK). These facilities stand for more than 90% of the anthropogenic input since the start of the nuclear era during the early 1940's.

The atmospheric chemistry of iodine is rather complex and is associated with many active oxidation-reduction transformations, which are also referred to as the “iodine active mobile pool”. The complex chemistry together with absence of systematic data on iodine in the continental atmosphere has contributed to uncertainties in establishing transport pathways of iodine from oceans to land. The transport may be dominated by fine-particles (<1 microm organo-salts) and/or agglomeration of gaseous molecules (Baker et al., 2000). Whether the transport occurs via wet or dry modes is controversial,

but recent data on aerosols suggest that dry transport can be less dominant mechanism compared to the wet one (Truesdale and Jones, 1996; Englund et al. 2008). Precipitation (wet deposition) would also be the major media that brings iodine to the Earth's surface, but whether all the iodine is cloud-bound or partly below cloud wash out is uncertain.

High latitude regions are very sensitive to anthropogenic contamination and together with the wide latitudinal coverage of Scandinavia, low aerosol loading, variable air mass sources and proximity to different marine systems make the region ideal for a case study. Preliminary investigations about iodine distribution in North Europe and in particular the Baltic Sea region and Sweden, clearly demonstrated strong enhancement (100-10000) in the  $^{129}\text{I}$  concentrations above the natural values in the environment. This conclusion together with the insignificant releases from the nuclear power plants in Scandinavia and the Baltic States (Hou et al., 2002) point out the remote discharges as the main sources.

Putting in mind a near future increase in  $^{129}\text{I}$  releases from the nuclear energy industry, there is indispensable need for expanding our knowledge about atmospheric dispersal and transport pathways of the isotope. Under such premises Scandinavia, and most likely other parts of the world, will eventually receive more  $^{129}\text{I}$  fallout. Although the fallout will be in both dry and wet modes, experience from the Chernobyl accident, clearly shows that extent of anthropogenic fallout can strongly be dependent on precipitation events. Thus we targeted mainly precipitation in our report here. The study represents a first evaluation of  $^{129}\text{I}$  and  $^{127}\text{I}$  in the precipitation (rain and snow) covering a period of 7 years and a latitude range from 55°N-68°N. About 90% of the precipitation

samples were collected during single events and thus would reflect contemporary atmospheric iodine fallout.

## **2. Samples and analytical techniques**

Precipitation samples used in this study were collected from three sites in Sweden, and one site in Denmark (Table 1 and Fig. S1). Normally, precipitation events were immediately collected, but in some cases more than one sample was taken from the same precipitation event. The sampling period may vary from <1 hour to over a few days. Monthly precipitation samples were collected for the Danish site.  $^{129}\text{I}$  was extracted from water samples for accelerator mass spectrometry (AMS) measurements. Measurement of  $^{127}\text{I}$  was performed at the Risø National Laboratory in Denmark using X SeriesII (Thermo Scientific) inductively coupled plasma mass spectrometer (ICP-MS). Detailed description of sampling sites, sampling procedure, analytical techniques and results (Table S1 and figures S1-S6) are given in the supplementary information.

## **3. Results**

Summary of  $^{129}\text{I}$  and  $^{127}\text{I}$  data are presented in Table 1 and the sampling sites will be hereafter designated by their latitude. The overall range of  $^{127}\text{I}$  concentration is 0.05-10.8  $\mu\text{gL}^{-1}$  with the highest values occurring in rain at site 60°N and the lowest in snow at site 68°N. Snow from site 56°N shows the lowest average concentration (0.49  $\mu\text{gL}^{-1}$ ) compared to the other sites.

However, we observe a clear geographic dependence in the average values of  $^{127}\text{I}$  between the southernmost sites (55°N and 56°N) and the most northern site (68°N). Samples from site 55°N represent monthly averages of only one year and thus reflect

lower statistical confidence. The  $^{127}\text{I}$  data do not show significant inter-and intra-annual variability, but generally more iodine occurs in the spring-summer precipitation (rain) compared to the autumn-winter (snow) season (Figs. S2-S4). Unlike  $^{127}\text{I}$ , the range of variation in the  $^{129}\text{I}$  concentration is wider between sites with respect to individual precipitation ( $0.4\text{-}298 \times 10^8 \text{ atoms L}^{-1}$ ) and averages ( $7.4\text{-}42 \times 10^8 \text{ atoms L}^{-1}$ ) (Table 1). As is the case with  $^{127}\text{I}$ , there are clear differences in the  $^{129}\text{I}$  between the southern and northern sites and higher concentrations are also more frequent in the rain compared to snow (Figs. S2-S4). Compared to data from Europe, the  $^{127}\text{I}$  and  $^{129}\text{I}$  concentrations shown in Table 1 are within the reported values in precipitation (Persson et al. 2007).

The annual  $^{127}\text{I}$  and  $^{129}\text{I}$  fluxes range at  $0.34\text{-}1.5 \text{ mg m}^{-2} \text{ y}^{-1}$  and  $3\text{-}30 \times 10^{11} \text{ atoms m}^{-2} \text{ y}^{-1}$  with average values of about  $1.0 \text{ mg m}^{-2} \text{ y}^{-1}$  and  $15 \times 10^{11} \text{ atoms m}^{-2} \text{ y}^{-1}$  respectively. These average fluxes result in annual  $^{129}\text{I}$  and  $^{127}\text{I}$  deposition on northern Europe (Denmark, Sweden, Norway and Finland) at about 60g and  $1.2 \times 10^9 \text{ g}$  respectively. The average annual  $^{129}\text{I}$  wet deposition represent  $< 1\%$  and  $< 0.05\%$  of the total annual gaseous and liquid, respectively, discharges from the Sellafiled and La Hague Facilities. The  $^{127}\text{I}$  annual wet deposition represents less than 1% of the estimated global oceanic iodine flux, which is estimated at about  $10^{11}\text{-}10^{12} \text{ g y}^{-1}$  (Bell et al., 2002; O'Dowd et al., 2002)).

Variability in the isotopic ratio ( $^{129}\text{I}/^{127}\text{I}$ ) indicates a range of  $(0.1\text{-}29) \times 10^{-7}$  and averages at  $(1.6\text{-}4.9) \times 10^{-7}$ . The lowest and highest values are found at site 60°N without specific trends between rain and snow or during the year (Figs. S2-S4). Only a few data point show extremely high  $^{129}\text{I}/^{127}\text{I}$  values and occur in all sites investigated which is also reflected by the range of the average values.

All the stations have peaks in the stable iodine concentrations in spring and autumn and lower concentration during the summer and especially winter months. A possible explanation for the low winter concentrations, apart from the fact that snow seems to be less inclined to incorporate iodine compounds, is that much of Sweden is covered with snow during winter, which can prevent re-suspension of iodine from the ground. An explanation of the lower summer concentrations could be a higher frequency of local rain events from large lakes and the Baltic Sea as the main moisture sources. Accordingly, the weak seasonal differences observed in the studied sites is different from the situation reported in the U.K. where no seasonal dependence of iodine in precipitation is found (Truesdale and Jones, 1996; Baker et al., 2001).

## **5. Discussion**

The transfer mechanisms of iodine from oceans into the atmosphere are not fully understood, and can depend on solar radiation, ocean water temperature and chemistry, biological activity (micro- and macro-algae), physical parameters (waves, tides and currents), and spatial extent (open sea/coastal areas and water depth) (Bell et al., 2002 and references therein). Once in the atmosphere, iodine exists in many forms, several of which undergo photolytic reactions quite fast. Despite this complex behavior of iodine, an attempt is presented here to elucidate some of the patterns observed in the results of this study. As mentioned in the introduction >90% of the  $^{129}\text{I}$  inventory is related to releases from the nuclear reprocessing facilities at Sellafield and La Hague. The releases are in liquid (discharged into the sea) and gaseous (discharged into the atmosphere) forms. Gaseous releases can be transported to Sweden through dispersion with westerly winds (prevailing wind direction at these latitudes) in dry (aerosols and gaseous form)



and wet (moisture and clouds) modes. Liquid releases can be transferred to the marine atmosphere and transported to Sweden in both dry and wet modes. Dry deposition is suggested to represent about 50-60% of the total iodine deposition particularly at large aerosol particle size ( $>1 \mu\text{m}$ ) (Baker et al., 2001), but the data by Englund et al. (2008) suggest  $<25\%$  as a contribution by dry deposition in Sweden. However, wet deposition can include washout of aerosols and gaseous iodine from the atmosphere, and hence would give reasonable recovery of the total iodine concentration in the atmosphere. Such an indication is also manifested by samples from the later phase of some precipitation events, where lower concentrations are observed in end than in the beginning of the same event, thus reflecting scavenging of dry mode (aerosols and gas) from the atmosphere.

The trend of, generally, lower iodine concentrations in precipitation of site  $68^\circ\text{N}$  compared to  $56^\circ\text{N}$  (Table 1; Fig. S2-S4) may partly relate geographical location, where sheltering by the Scandinavian mountains would enhance contribution from local moisture sources compared to long distance ocean-related transport. Indications of effects on iodine transport along low topography landscape (boundary layer effect; Saiz- Lopez et al., 2006) and less incorporation in high altitude snow (Gilfedder et al., 2007) also verify our observations. On the other hand, proximity of sites  $56^\circ\text{N}$  and  $55^\circ\text{N}$  to marine waters (North Sea and Skagerrak/ Kattegat) can explain the high iodine concentrations. Iodate ( $\text{IO}_3$ ) is the most common form of iodine in precipitation, but the relative availability of iodide ( $\text{I}^-$ ) and  $\text{I}_2$  in the atmosphere can depend on proximity to marine waters (Campos et al., 1996; Truesdale and Jones, 1996). The later iodine species are relatively metastable compared to iodate and thus can be removed from the atmosphere and accordingly reduce the iodine concentration with further distance from the sea. The

effect of proximity to marine waters is also reflected by the difference in the average iodine concentration between site 60°N and 55°-56°N. The earlier site is more close to the Baltic Sea (Fig. S1), which has > 50% less concentration of  $^{127}\text{I}$  and about 2 orders of magnitude less concentration of  $^{129}\text{I}$  than the North Sea and Skagerrak/Kattegat (Truesdale et al., 2001; Aldahan et al., 2007; Hou et al., 2002, 2007). Apart from the varying concentrations in source regions, precipitation rates may also influence the annual deposition. The annual precipitation at sites 55°N and 56°N is about twice as much as in 68°N (Table 1), which will enhance the spatial differences in annual deposition compared to average concentrations.

A factor that may cause different distributions of  $^{127}\text{I}$  and  $^{129}\text{I}$  in precipitation is their respective sources to the atmosphere. Although both isotopes can have marine waters as the main source,  $^{129}\text{I}$  has specific point sources related to the discharges from the Sellafield and La Hague facilities. In addition to the liquid discharges, which are dominating the  $^{129}\text{I}$  marine inventory, there is a substantial amount of gaseous releases that can influence the atmosphere of North Europe. In order to estimate possible main sources of marine waters and contributions from gaseous releases, we have compared concentrations and isotope ratios in precipitation with corresponding values for the ocean. For stable iodine, the precipitation/ocean concentration ratio is about 1/50. The same ratio should be true for the radioactive isotope, which implies ocean concentrations of  $5 \times 10^{10}$  atoms  $\text{L}^{-1}$  to account for a concentration of  $1 \times 10^9$  atoms  $\text{L}^{-1}$  in precipitation. Such high concentrations are observed in the North Sea and Skagerrak/Kattegat, (Alfimov et al., 2004a; Hou, 2004), whereas concentrations in the surface waters of the North Atlantic ( $10^7$ - $10^8$  atoms  $\text{L}^{-1}$  [Alfimov et al., 2004a]), the Baltic Sea and Barents Sea ( $10^9$  atoms  $\text{L}^{-1}$

<sup>1</sup> [Alfimov et al., 2004b; Aldahan et al., 2007]) cannot account alone for the relatively high concentrations found in the measured precipitation. Isotopic ratio in precipitation is  $1-5 \times 10^{-7}$  (Table 1), whereas isotope ratios in the ocean water of the North Atlantic are in the order of  $10^{-9}$  (Alfimov et al., 2004a, Hou, 2004). In the North Sea and the Skagerrak/Kattegat the  $^{129}\text{I}/^{127}\text{I}$  values are  $10^{-7} - 10^{-6}$  (Hou et al., 2008). Accordingly, contribution from waters with higher  $^{129}\text{I}$  concentrations than in the ones found in the North Atlantic and/or direct gaseous emission are needed to account for the high isotopic ratio in precipitation. Since clouds normally contain moisture from a large area, precipitation samples with extreme high ratios ( $>10^{-6}$ ) are probably influenced by the gaseous releases. However, the low temporal resolution of the available discharge data (1 month; Fig. S5) hinders accurate analysis of the impact of gaseous releases on  $^{129}\text{I}$  in precipitation.

Presently, we have no data about anthropogenic sources of  $^{127}\text{I}$  within North Europe, but fossil fuel and biomass burning may provide insignificant amounts compared with natural sources (ocean, wetlands, soil and vegetation). Thus we suggest that proximity to marine coastal environment may explain the geographic difference in the isotope concentration, being higher at the southern compared to northern sites. The major carrier compound of iodine from the ocean and land to the atmosphere is methyl iodide ( $\text{CH}_3\text{I}$ ), although several other compounds ( $\text{CH}_2\text{I}_2$ ,  $\text{CH}_3\text{-CH}_2\text{I}$ ,  $\text{CH}_2\text{-CH}$ ,  $\text{HOI}$ ) and also molecular  $\text{I}_2$  and particulate iodine may be emitted and form during the journey from the source to the sink (precipitation). With such a complex chemical and physical pattern of iodine occurrence and transport in the atmosphere, achieving a decisive conclusions on transport pathways represent a tough challenge. Accordingly, information from

combining both isotopes can provide better understanding of the iodine atmospheric pathways. Weak positive correlation (0.6) between  $^{127}\text{I}$  and  $^{129}\text{I}$  (Fig. S6) indicates that these two isotopes are partly decoupled mainly due to different source functions and transport modes. The trapping of iodine molecules in the rain droplets may relate to equilibrium kinetics between the two isotopes, which may reflect speciation modes in the atmosphere. Also, despite a clear geographic difference in the distribution of the isotopes (being higher in the near coast site), there is no coherent pattern with respect to magnitude of change in the isotopes concentrations (Figs. S2-S4). This behavior can be explained by differences in expected chemical forms and transport pathways of the two isotopes. If iodine isotopes do have different chemical forms in precipitation, then that must stem from source contribution. Causes behind this speciation difference may relate to: 1) Effect of the regional and local sources and 2) Dominant speciation of the emitted iodine molecules from the source. The major form of  $^{129}\text{I}$  that is emitted through gaseous and liquid sources is iodide, and together with persistent or slow conversion rate of the iodide species in the atmosphere and marine water (Baker et al., 2001; Hou et al. 2008) suggest possible high proportion of  $^{129}\text{I}$  as iodide in precipitation.

One important parameter that helps understanding the varying levels of  $^{127}\text{I}$  and  $^{129}\text{I}$  in precipitation is origin of the air mass causing the precipitation event. We have reconstructed the air masses pathways for most of the precipitation events sampled in this study using the HYSPLIT (HYbrid Single- Particle Lagrangian Integrated Trajectory) model developed in cooperation between NOAA (U.S National Oceanic and Atmospheric Administration) and the Australian Bureau of Meteorology. Detailed description of the model is published in Draxler and Hess (2004) and the one used here is

given in the supplementary information.

Back trajectories were calculated through assigning a “region of origin” for each sampled precipitation event (Fig. S1). An example of back trajectories starting at site 56°N and passing over the Sellafield/La Hague area is shown in Figure 1 and a summary of the data is presented in Table S1. The average  $^{129}\text{I}$  concentration in region 1 (Sellafield-La Hague and North Sea) shows elevated level when compared to the other regions for all sites.

Additionally, all samples from 56°N with an isotope ratio  $>4 \times 10^{-7}$  (8 samples) are found in region 1. This pattern indicates a connection between extreme high values in site 56°N and air masses passing over the Sellafield/La Hague area.

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Table 1. Summary data of precipitation in the sites used and other regions.

Fig. 1. Example of a back trajectory starting at site 56°N at 20:00 hour on June 30, 2001 and continued at a three hours interval (backward in time) for different altitudes. Precipitation associated with the air masses has passed through Sellafield and La Hague region. The lower part of the figure shows the changes in the vertical movement of the air mass trajectories.



Site	Altitude m.a.s	Annual ppt (mm)	Sampling	<sup>127</sup> I (microg/L)		<sup>127</sup> I (mg/m <sup>2</sup> /y)	<sup>129</sup> I (10 <sup>8</sup> atoms/L)		<sup>129</sup> I (10 <sup>11</sup> atoms/m <sup>2</sup> /y)	<sup>129</sup> I/ <sup>127</sup> I (10 <sup>-7</sup> atom/atom)	
				range	average		range	average		range	average
68.35°N 18.81°E	385	330	Dec 2000-Nov 2005 P R=73 (55) S=68 (54)	0.05-6.1 0.11-6.1 0.05-2.0	0.83 1.05 0.59	0.34	0.4-61 1.3-61 0.4-32	8.9 9.1 7.4	3.0	0.6-18 1.1-10 0.8-18	2.5 2.7 2.4
59.85°N 17.63°E	25	545	Apr 2000-Feb 2006 P R= 123 (76) S= 71 (52)	0.13-10.8 0.18-10.8 0.13-4.0	1.83 2.37 1.05	1.2	0.7-82 1.1-55 0.7-82	11 12 8.0	6.0	0.1-29 0.2-4.6 0.1-29	1.6 1.2 2.2
56.13°N 13.05°E	30	700	June 2001-Dec 2005 P R=53 (23) S=9 (9)	0.12-9.3 0.76-9.3 0.12-1.1	2.13 2.77 0.49	1.5	1.4-298 9.2-298 1.4-39	38 42 5.8	30	0.8-27 0.9-27 0.8-11	4.1 4.0 4.5
55.69°N 12.10E	25	560	Jan-Dec 2000 P=12 <sup>1</sup>	1.3-2.8	1.83	1.0	10-85	40	22	1.4-11	4.9
Europe <sup>2</sup>			Total precipitation	0.3-10			0.3-600				
World <sup>3</sup>			Total precipitation	0.2-12			0.01-600				

1 monthly average

2 From Person et al. (2007)

3 Based on data from the Northern Hemisphere (Truesdale and Jones 1996; Aldahan et al. 2007)

P= total precipitation, R= rain, S= snow

Samples analysed for <sup>127</sup>I are marked between parentheses

Table 1.

## NOAA HYSPLIT MODEL

Backward trajectories ending at 24 UTC 30 Jun 01

FNL Meteorological Data

