



Speciation of  $^{127}\text{I}$  and  $^{129}\text{I}$  in atmospheric aerosols at Risø

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# Speciation of $^{127}\text{I}$ and $^{129}\text{I}$ in atmospheric aerosols at Risø, Denmark: insight into sources of iodine isotopes and their species transformations

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## Abstract

Speciation analysis of iodine in aerosols is a very useful approach for understanding geochemical cycling of iodine in the atmosphere. In this study, overall iodine species, including water-soluble iodine species (iodide, iodate and water-soluble organic iodine), NaOH-soluble iodine and insoluble iodine have been determined for  $^{129}\text{I}$  and  $^{127}\text{I}$  in the aerosols collected at Risø, Denmark, between March and May 2011 (shortly after the Fukushima nuclear accident) and in December 2014. The measured concentrations of total iodine are in the range of  $1.04\text{--}2.48\text{ ng m}^{-3}$  for  $^{127}\text{I}$  and  $(11.3\text{--}97.0) \times 10^5\text{ atoms m}^{-3}$  for  $^{129}\text{I}$ , and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios of  $(17.8\text{--}86.8) \times 10^{-8}$ . The contribution of Fukushima-derived  $^{129}\text{I}$  (peak value of  $6.3 \times 10^4\text{ atoms m}^{-3}$ ) is estimated to be negligible (less than 6 %) compared to the total  $^{129}\text{I}$  concentration in northern Europe. The concentrations and species of  $^{129}\text{I}$  and  $^{127}\text{I}$  in the aerosols are found to be strongly related to their sources and atmospheric pathways. Aerosols that were transported over the contaminated ocean, contained higher amounts of  $^{129}\text{I}$  than aerosols transported over the European continent. The high  $^{129}\text{I}$  concentrations of the marine aerosols are attributed to secondary emission from heavily  $^{129}\text{I}$ -contaminated seawater rather than primary gaseous release from nuclear reprocessing plants. Water-soluble iodine was found to be a minor fraction to total iodine for both  $^{127}\text{I}$  (7.8–13.7 %) and  $^{129}\text{I}$  (6.5–14.1 %) in ocean-derived aerosols, but accounted for 20.2–30.3 % for  $^{127}\text{I}$  and 25.6–29.5 % for  $^{129}\text{I}$  in land-derived aerosols. Iodide was the predominant form of water-soluble iodine, accounting for more than 97 % of the water-soluble iodine. NaOH-soluble iodine seems to be independent of the sources of aerosols. The significant proportion of  $^{129}\text{I}$  and  $^{127}\text{I}$  found in NaOH-soluble fractions is likely bound with organic substances. In contrast to water-soluble iodine however, the sources of air masses exerted distinct influences on insoluble iodine for both  $^{129}\text{I}$  and  $^{127}\text{I}$ , with higher values for marine air masses and lower values for terrestrial air masses.

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## 1 Introduction

The atmospheric chemistry of iodine has attracted increasing attention in recent years, in part because of its central role in governing the geochemical cycling of iodine, and also for its influences on ozone depletion, primary particle formation, and global climate (Carpenter, 2003; O'Dowd et al., 2002; Saiz-Lopez et al., 2012). Most atmospheric iodine studies to date have concentrated on laboratory smog chamber experiments and modeling studies, with a paucity of field measurements of atmospheric samples. Apart from gaseous iodine (e.g. molecular iodine, iodocarbons) in the atmosphere, aerosols contain a significant fraction of iodine, and play an important role for iodine transportation from ocean to land. In particular, speciation analysis of iodine in aerosols is expected to speculate the reactions and atmospheric processes of iodine, and develop a comprehensive understanding of biogeochemical cycles of iodine in nature.

Most of the work on aerosol iodine speciation has focused on water-soluble iodine (WSI) (Baker, 2004, 2005; Gilfedder et al., 2008). Early modeling studies suggested that aerosol iodate should be the only stable iodine specie as a by-product of iodine oxides production, with negligible iodide due to transformation into gaseous iodine (McFiggans et al., 2000; Vogt et al., 1999). However, this prediction went against several previous observations that showed significant iodide and soluble organic iodine, accounting for 10–100% of WSI in aerosols (Baker, 2004, 2005; Gabler and Heumann, 1993). An improved model was then developed, primarily by increasing the proportion of iodide in the aerosols (Pechtl et al., 2007). Few studies have considered water-insoluble iodine in aerosols (Gilfedder et al., 2010; Tsukada et al., 1987), which might hardly be converted to gaseous iodine before being deposited on the Earth's surface. Existing observational data suggest that water-insoluble iodine is abundant in aerosol particles, representing 17–53% of the total iodine (Gilfedder et al., 2010). However, there is no comprehensive investigation of aerosol iodine species that encompasses aqueous-soluble and insoluble iodine species. This constitutes a significant gap in our understanding of the interaction among atmospheric iodine species.

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Due to the difficulty of source identification of natural  $^{127}\text{I}$ ,  $^{129}\text{I}$  ( $T_{1/2} = 15.7\text{ Ma}$ ) has proven to be an especially useful geochemical tracer both in the marine environment and in the atmosphere (Hou et al., 2007; Jabbar et al., 2012; Michel et al., 2012). A large amount of  $^{129}\text{I}$  has been released to the environment by nuclear reprocessing plants (NRPs). These dominate all other anthropogenic sources, with 6000 kg released as of 2009, from Sellafield (United Kingdom) and La Hague (France) (He et al., 2013b; Hou et al., 2007; Raisbeck et al., 1995). Anthropogenic  $^{129}\text{I}$  is currently distributed unevenly around the globe in water, atmosphere and terrestrial systems (Hou et al., 2009b). Above-ground nuclear explosions also produced  $^{129}\text{I}$ , injecting it into the troposphere and stratosphere. The proportion that entered to the stratosphere became mixed globally before re-entering the troposphere. Larger particles remained in the troposphere for about 20 days before being deposited on the earth's surface (Tölggyessy, 1993). Gaseous releases from reprocessing plants and accidents are primary sources of  $^{129}\text{I}$  to the local environment (Hou et al., 2009a; Xu et al., 2013). Re-emission of iodine from the marine boundary layer plays a significant role in the atmospheric budget of  $^{129}\text{I}$  (Englund et al., 2010b). As with stable iodine ( $^{127}\text{I}$ ),  $^{129}\text{I}$  also travels in the atmosphere in particulate form. Particulate  $^{129}\text{I}$  has been used successfully in monitoring studies, and as an atmospheric tracer (Englund et al., 2010b; Jabbar et al., 2012; Santos et al., 2005; Tsukada et al., 1987). While, speciation analysis of  $^{129}\text{I}$  in aerosols is extremely scarce with only one of our previous study (Xu et al., 2015).

Here, we present the results of speciation analysis of stable  $^{127}\text{I}$  and radioactive  $^{129}\text{I}$  in aerosols collected in Denmark. Our goal is to understand how the iodine is carried in the atmosphere and to investigate transformation mechanisms between iodine species in aerosols, as well as gas-aerosol exchange processes involving iodine. To our knowledge, this is the first paper to address the conversion among overall iodine species in aerosols.





during 1983–2008 (Englund et al., 2010b; Michel et al., 2012). However, the measured  $^{129}\text{I}$  concentrations and  $^{129}\text{I}/^{127}\text{I}$  ratios are 1–2 orders of magnitude higher than those observed in Vienna, Austria during 2001–2002, and in the high altitude eastern Alps (2700 m) during 2001 (Jabbar et al., 2011, 2012).

For  $^{127}\text{I}$ , the WSI fraction of aerosols collected in 2011 occurred predominantly as iodide, ranging from 0.12–0.33  $\text{ng m}^{-3}$ . Minor iodate of 0.02–0.03  $\text{ng m}^{-3}$  was detected only in two samples (AE11–12 during 4–7 April and AE11–17 during 26 April–2 May) (Table 1 and Fig. 4). The highest measured  $^{127}\text{I}^-$  concentration (0.74  $\text{ng m}^{-3}$ ) was observed in aerosol collected in 2014, and there was no measurable water-soluble organic iodine. Water-soluble  $^{129}\text{I}$  shows a similar species pattern as water-soluble  $^{127}\text{I}$  except that iodate-129 was below the detection limit in all the analyzed samples. The concentrations of  $^{129}\text{I}^-$  varied in the range of  $(3.26\text{--}5.91) \times 10^5 \text{ atoms m}^{-3}$  with an average value of  $4.41 \times 10^5 \text{ atoms m}^{-3}$  in the aerosols from 2011. This value is about 8 times lower than that ( $30.12 \times 10^5 \text{ atoms m}^{-3}$ ) in the sample collected in 2014. High NSI concentrations were observed in all of the aerosol samples, with an average of  $0.64 \pm 0.21 \text{ ng m}^{-3}$  for  $^{127}\text{I}$  and  $(13.55 \pm 10.12) \times 10^5 \text{ atoms m}^{-3}$  for  $^{129}\text{I}$ . RII is the dominant species in aerosol samples for both iodine isotopes, with concentration range of  $0.34\text{--}1.66 \text{ ng m}^{-3}$  for  $^{127}\text{I}$  and  $(4.27\text{--}39.94) \times 10^5 \text{ atoms m}^{-3}$  for  $^{129}\text{I}$ .

There is no available standard reference material for iodine isotopes in aerosols which can be used for validation of the analytical method. While, the sums of all of iodine species in the analyzed samples are comparable with the measured total iodine in the corresponding samples for both  $^{127}\text{I}$  and  $^{129}\text{I}$  in a range of 85–110 %, indicating the reliability of our analytical results (Fig. 5).

For the samples collected before 18 April 2011, the proportion of  $^{129}\text{I}$  and  $^{127}\text{I}$  species follows an order of  $\text{RII} > \text{NSI} > \text{iodide}$ . For the samples collected later in April 2011 and in the winter 2014, the three iodine fractions for both  $^{129}\text{I}$  and  $^{127}\text{I}$  account for almost equivalent proportion of total iodine (Fig. 5). Compared to the former sampling period (31 March–18 April 2011), the most apparent feature of the latter sampling period is a dramatic diminution of iodine in the RII fraction, by 38.7 % for  $^{127}\text{I}$  and 26.8 % for  $^{129}\text{I}$ .

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This was compensated by a corresponding increase in the iodide fraction of the aerosol particles. Iodate, as the least abundant iodine specie (< 3 %), was only observed in two aerosol samples for  $^{127}\text{I}$ , and no  $^{129}\text{IO}_3^-$  was detected.

An range of  $^{129}\text{I}/^{127}\text{I}$  atomic ratios of  $(15.56\text{--}102.36) \times 10^{-8}$  was determined for the different iodine species in aerosols. The variations of  $^{129}\text{I}/^{127}\text{I}$  ratio in iodide, NSI and RII are similar to those in total  $^{129}\text{I}/^{127}\text{I}$ , with high values observed prior to 18 April 2011 and December 2014 (Table 2 and Fig. 3).

## 4 Discussion

### 4.1 Sources of $^{127}\text{I}$ and $^{129}\text{I}$ in aerosols

Concentration variations of  $^{127}\text{I}$  and  $^{129}\text{I}$  in aerosols against meteorological parameters (i.e. wind direction, wind speed and temperature) during the sampling period show that wind direction has a dominant influence (Fig. 3). Back trajectory model analysis shows that  $^{127}\text{I}$  and  $^{129}\text{I}$  in the aerosols was directly controlled by the sources and pathways of air masses (Figs. 6, S1 and S2 in the Supplement). The relatively high  $^{127}\text{I}$  and  $^{129}\text{I}$  concentrations was observed in in aerosols collected early April 2011 and December 2014, when the air mass was mainly transported from the Atlantic Ocean and the North Sea by prevailing westerly winds. Relatively low concentrations of iodine isotopes were observed in the aerosols collected later in April, when the air masses were dominated by prevailing easterly winds and passed over the European continent and the Baltic Sea.

Marine emission of volatile iodine species is a major source of iodine in the atmosphere. It results in relatively elevated  $^{127}\text{I}$  concentrations in the marine atmosphere, as compared to the terrestrial atmosphere (Saiz-Lopez et al., 2012). During the sampling period of 11–14 April, the sampled air mass was transported by westerly winds from a vast area over the northern Atlantic Ocean. This caused an elevated  $^{127}\text{I}$  concentration. Except for the Norwegian Sea, most areas of the northern Atlantic have low seawater

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aerosol formation. In contrast, iodine in precipitation originates from washout process of gaseous iodine and air-borne particulate iodine, primarily in the form of polar iodine compounds (e.g. HI, HOI, IO, I<sup>-</sup>, IO<sub>3</sub><sup>-</sup>, I<sub>2</sub> and unidentified organic iodine species) in H<sub>2</sub>O molecules during precipitation events (Buraglio et al., 2001; Gilfedder et al., 2007). In addition, aerosol samples were collected over a relatively long time scale (3–7 days), reflecting an accumulated signal during the sampling period, while precipitation events are normally much shorter. Therefore, the source terms of iodine isotopes in precipitation might not necessarily follow back trajectory analysis.

## 4.2 Species of <sup>129</sup>I and <sup>127</sup>I in aerosols

WSI is virtually pure iodide in the aerosols investigated, with iodate and water-soluble organic iodine accounting for less than 3% of total iodine, and these are only measurable in two samples. Iodate was once considered to be the only WSI species in aerosol (Vogt et al., 1999). This was supported by earlier field observations demonstrating that iodate was dominant in size-segregated aerosols from the tropical Atlantic (Wimschneider and Heumann, 1995). However, this iodate-dominant feature was not found in other aerosol samples, e.g. in the northwest Atlantic Ocean and in tropical atmospheric aerosols (Baker, 2004, 2005). In these cases, iodide was the dominant aerosol phase, as observed in this study. Significant amounts of soluble organic iodine, accounting for 83–97% of WSI, has been reported in aerosols collected at the Mace Head atmospheric research station on the west coast of Ireland (Gilfedder et al., 2008). Water-soluble organic iodine accounting for 4–75% of WSI were also measured in aerosols collected from a cruise from the UK to the Falkland Islands in 2003 (Baker, 2005). This suggests that the proportion of soluble organic iodine in aerosols varies regionally and depends on particular aerosol sources and formation processes. Some of this variability might also be related to the analytical methods employed for speciation analysis (Zhang et al., 2015).

It is not clear how iodide is formed in the atmosphere, in an oxidizing environment containing oxygen and ozone. Early models predict a negligible iodide concentration in

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particle phases based on an assumption that the iodide in aerosols only originates from the low levels of gaseous HI (McFiggans et al., 2000; Vogt et al., 1999). This work in combination with other previous results (Baker, 2004; Xu et al., 2015) suggests that there must be alternative primary pathways that contribute to iodide formation at the observed levels. It's generally accepted that iodine atoms are photochemically produced by photolysis of gaseous iodinated compounds, and oxidized by ozone to form reactive iodine oxides (Carpenter, 2003; Saiz-Lopez et al., 2012; Vogt et al., 1999). The formation of iodide from iodine atoms and other reactive iodine compounds must rely on electron-donors that are capable of reducing high valence iodine species to iodide. One possibility is the involvement of sulfur compounds (Chatfield and Crutzen, 1990). Possible reaction pathways are given in Table 3. Gaseous  $\text{SO}_2$  can be formed in nature by reactions of dimethyl sulfate (DMS) with hydroxide and nitrate. By associating with  $\text{H}_2\text{O}$ , these reactions produce  $\text{HSO}_3^-$  and  $\text{SO}_3^{2-}$  (Eqs. 1 and 2). Native iodine and other reactive species (not shown) can be reduced to  $\text{I}^-$  on gas-aerosol interfaces (Eq. 3). Other iodine species in aerosols can also be reduced by reductive sulfur compounds to form iodide (Eq. 4). The electron-donors are not limited to sulfur compounds either, for example, nitrogen in the form of gaseous ammonia ( $\text{NH}_3 \rightarrow \text{NO}_2/\text{NO}_3$ ) (McFiggans et al., 2000; Saiz-Lopez and Plane, 2004) and elemental mercury ( $\text{Hg}^0 \rightarrow \text{HgO}/\text{HgX}$ , where  $X$  is a halogen,  $\text{I}^-$ ,  $\text{Br}^-$  or  $\text{Cl}^-$ ) (Lindberg et al., 2002; Simpson et al., 2007) are also candidates responsible for iodide formation.

We note that relatively low WSI  $^{129}\text{I}$  and  $^{127}\text{I}$  was measured in marine-sourced aerosols from the North Sea, as compared to the European continent-sourced aerosols (Figs. 4 and 5). This is consistent with the findings drawn from an iodine speciation study of coastal aerosol samples from England (Baker et al., 2001), where the concentrations of total water-soluble iodine from continental aerosols were significantly higher than those from marine aerosols.

A large proportion of  $^{129}\text{I}$  and  $^{127}\text{I}$  in our aerosol samples were NaOH-soluble, which is consistent with the results of aerosol from Tsukuba, Japan, collected shortly after the Fukushima nuclear accident in March 2011 (Xu et al., 2015). Abundant NaOH-soluble



releasing  $^{129}\text{I}$  into the European environment for about 50 years, allowing it to follow geochemical pathways on timescales ranging from days to decades. In contrast, RII from Fukushima-derived aerosols had only days to react with their environment prior to sampling, 15–22 March 2011.

The origin of the RII fraction is not well understood at present. It's possible that the RII fraction is derived from suspended soil particles (Xu et al., 2013). However, speciation analysis of  $^{129}\text{I}$  in soils show that residual iodine after NaOH leaching accounts for less than 10 % of the total, and this minor component is assumed to be associated with minerals (Hou et al., 2003; Qiao et al., 2012). Another possibility is a connection with soot and black carbon from combustion processes, found in anthropogenic aerosols (Kondratyev et al., 2006; Rose et al., 2006). However, the aerosols collected for this study in early April 2011 and winter of 2014 were mainly marine aerosols, with high RII. Alternatively, the RII fraction in aerosols might be associated with metal oxides that originated by suspension of fine inorganic particles. A relatively large fraction of iodine in soil and sediment has been observed in metal oxides associated form (Hou et al., 2003). Gaseous iodine in the atmosphere could interact with these inorganic particles and become firmly bound. The relatively high RII in the marine aerosols (Figs. 4 and 5) suggests that some marine components facilitate the association of gaseous iodine with inorganic particles.

A significant positive correlation between  $^{127}\text{I}$  iodide and  $^7\text{Be}$  in aerosol samples ( $R = 0.76$ ,  $p = 0.05$ ) is observed (Fig. 7).  $^7\text{Be}$  ( $t_{1/2} = 54$  days) is produced in the stratosphere by cosmic ray reactions with oxygen and nitrogen and rapidly transported to the troposphere by vertical mixing processes to be finally deposited onto the earth. The production of  $^7\text{Be}$  decreases with decreasing altitude in the troposphere because of the attenuation of cosmic rays by interactions with atomic targets in the atmosphere (Turekian et al., 1983). This positive correlation might indicate that the formation of iodide occurs in air masses at higher altitude. However, no significant correlation between  $^{129}\text{I}^-$  and  $^7\text{Be}$  is observed (Fig. 7). This is consistent with the process of secondary emission of  $^{129}\text{I}$  from contaminated seawater, which would only increase  $^{129}\text{I}$  in

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the troposphere. The significant negative correlation of NSI of  $^{129}\text{I}$  with  $^7\text{Be}$  ( $R = 0.73$ ,  $\rho = 0.06$ ) likely reflects the association of iodine with organic matter at low altitude, where organic matter is liberated by biological activity in the ocean and on land.

### 4.3 Fukushima-derived $^{129}\text{I}$ signal in the European atmosphere

5 The Fukushima Dai-ichi nuclear power plant accident on 11 March 2011 released radiiodine to the atmosphere, primarily as  $^{131}\text{I}$  and  $^{129}\text{I}$ , which was mainly transported eastwards by prevailing winds. Based on  $^{129}\text{I}$  levels in the offshore Fukushima seawater, the released  $^{129}\text{I}$  amount from this accident was estimated to be 1.2 kg (Hou et al., 2013). Gaseous  $^{129}\text{I}$  travelled across the Pacific Ocean, American continent and  
10 Atlantic Ocean, and some fraction arrived on the European continent after 1–2 weeks (Clemenza et al., 2012; Leon et al., 2011; Manolopoulou et al., 2011; Pittauerová et al., 2011). Anthropogenic  $^{129}\text{I}$  has been reported from a variety of environmental samples in Japan, including soil, seawater, precipitation and aerosols (Buessler et al., 2012; Hou et al., 2013; Muramatsu et al., 2015; Xu et al., 2013, 2015). The level  
15 of  $^{129}\text{I}$  in aerosols collected in Tsukuba, about 170 km from the Fukushima Dai-ichi NPP, reached  $5 \times 10^8$  atoms  $\text{m}^{-3}$  (Xu et al., 2015). While the Fukushima-derived  $^{129}\text{I}$  in environmental samples outside of Japan was less well-characterized.  $^{131}\text{I}$  in the aerosol samples collected at Risø, Denmark, 10 days after the Fukushima accident have been observed (Fig. 8) (Nielsen et al., 2011). The  $^{131}\text{I}$  reached the peak on 24–  
20 30 March 2011 ( $763 \mu\text{Bq m}^{-3}$  in aerosol), then fell to below detection limits for  $^{131}\text{I}$  in the middle of May. Based on the measured  $^{131}\text{I}$  radioactivity in the aerosol samples and an  $^{129}\text{I}/^{131}\text{I}$  atomic ratio of  $16.0 \pm 2.2$  deduced from the aerosol samples collected at Tsukuba, Japan shortly after the Fukushima accident (Xu et al., 2015), the Fukushima-derived  $^{129}\text{I}$  signal in Denmark can be reconstructed (Fig. 8). The highest  
25  $^{129}\text{I}$  concentration in the aerosols in Denmark from the Fukushima accident is estimated to be  $6.3 \times 10^4$  atoms  $\text{m}^{-3}$  on 30–31 March 2011, which accounts for less than  $\sim 6\%$  of total  $^{129}\text{I}$  ( $1.1\text{--}9.7 \times 10^6$  atoms  $\text{m}^{-3}$ ) in Denmark when the Fukushima  $^{131}\text{I}$  peak was

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measured. Considering the rapid decline of  $^{129}\text{I}$  levels in aerosols and precipitation in Japan to nearly pre-accident levels within two years (Xu et al., 2013), the contribution of Fukushima-derived  $^{129}\text{I}$  to the  $^{129}\text{I}$  level and inventory in Europe is now negligible in comparison to NRPs-derived  $^{129}\text{I}$  in the European atmosphere.

#### 5 4.4 Dry deposition flux of iodine isotopes

Dry deposition fluxes of iodine can be estimated as  $F_d = C_d \cdot v_d$  (Baker et al., 2001), where  $F_d$  is the flux ( $\mu\text{g m}^{-2} \text{ year}^{-1}$  for  $^{127}\text{I}$  and  $\text{atoms m}^{-2} \text{ year}^{-1}$  for  $^{129}\text{I}$ ),  $C_d$  is the aerosol iodine concentration in the atmosphere ( $\text{ng m}^{-3}$  for  $^{127}\text{I}$  and  $\text{atoms m}^{-3}$  for  $^{129}\text{I}$ ), and  $v_d$  is the deposition velocity ( $\text{cm s}^{-1}$ ). The deposition velocities are size-dependent, at  $0.1 \text{ cm s}^{-1}$  for fine particles (Aerodynamic diameter  $< 1 \mu\text{m}$ ) and  $1.5 \text{ cm s}^{-1}$  for larger particles (Duce et al., 1991). The aerosol samples in this work were collected on a PP filter with a pore size of approximately  $0.45 \mu\text{m}$ . According to the particle size distribution at Spieka-Neufeld, Germany, whose atmospheric input of iodine is also from the North Sea, as in this study, 67% of aerosol iodine accumulated in particles larger than  $1 \mu\text{m}$  and 33% in  $0.45\text{--}1 \mu\text{m}$  particles (Gabler and Heumann, 1993). Employing these parameters, dry deposition fluxes of iodine in Denmark can be estimated to be  $342\text{--}815 \mu\text{g m}^{-2} \text{ year}^{-1}$  for  $^{127}\text{I}$ , and  $(0.4\text{--}3.2) \times 10^{12} \text{ atoms m}^{-2} \text{ year}^{-1}$  for  $^{129}\text{I}$  during 2011–2014. If the average values of  $1.8 \text{ ng m}^{-3}$  and  $43.7 \times 10^5 \text{ atoms m}^{-3}$  are used as the representative concentrations of  $^{127}\text{I}$  and  $^{129}\text{I}$  in aerosol, respectively for 2011, average dry deposition fluxes of iodine in 2011 would be  $591 \mu\text{g m}^{-2} \text{ year}^{-1}$  and  $1.4 \times 10^{12} \text{ atoms m}^{-2} \text{ year}^{-1}$  for  $^{127}\text{I}$  and  $^{129}\text{I}$ , respectively. The  $^{127}\text{I}$  deposition flux falls within the range of natural stable iodine deposition, at  $460\text{--}830 \mu\text{g m}^{-2} \text{ year}^{-1}$ , observed at Norfolk, on the north coast of the United Kingdom (Baker et al., 2001). It is also comparable with measured values from southern Sweden, but higher than northern Sweden (Englund et al., 2010b). The dry deposition flux of  $^{129}\text{I}$  for aerosol samples collected in Sweden between 1983 and 2008 shows a range of  $0.33\text{--}6.6 \times 10^{11} \text{ atoms m}^{-2} \text{ year}^{-1}$  in southern Sweden, and

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the North Sea Norwegian coast water and Kattegat, which has been heavily contaminated by marine discharges of  $^{129}\text{I}$  from NRPs.

3. Fukushima-derived  $^{129}\text{I}$  contributed less than  $\sim 6\%$  to the total  $^{129}\text{I}$  in the European atmosphere immediately after the Fukushima accident, a negligible quantity when compared to  $^{129}\text{I}$  levels released from Sellafield and La Hague nuclear re-processing plants.
4. Water-insoluble iodine (NSI and RII) in aerosols that is primarily associated with organic compounds is crucial for investigation of geochemical cycling of iodine in the atmosphere due to its large fraction. This requires more intensive studies to extend our observations on both temporal and spatial scales.

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**Table 1.** Sampling information of aerosols collected at Risø, Denmark in 2011 and 2014. Data of  $^{131}\text{I}$ ,  $^7\text{Be}$  and  $^{210}\text{Pb}$  in the aerosol samples are adopted from the DTU Nutech report (Nielsen et al., 2011). The reference time was the mid-point of the sampling period, and analytical uncertainties were 5 % for  $^{131}\text{I}$ , and within 1 % for  $^7\text{Be}$  and  $^{210}\text{Pb}$ .

Sample No	Sampling date	Air flux, $\text{m}^3$	Air flux, $\text{m}^3 \text{h}^{-1}$	Weight, g	$^{131}\text{I}$ , $\mu\text{Bq m}^{-3}$	$^7\text{Be}$ , $\mu\text{Bq m}^{-3}$	$^{210}\text{Pb}$ , $\mu\text{Bq m}^{-3}$
AE11-1	31 Mar–04 Apr 2011	88 833	2757	72.5	205	1925	66
AE11-2	04–07 Apr 2011	64 339	2751	79.2	218	1482	47
AE11-3	07–11 Apr 2011	55 911	1744	79.5	147	1482	47
AE11-4	11–14 Apr 2011	27 083	1096	70.9	110	2750	172
AE11-5	14–18 Apr 2011	48 317	1505	77.9	58.3	2750	172
AE11-6	18–26 Apr 2011	101 400	1593	80.8	20.9	4528	249
AE11-7	26 Apr–02 May 2011	54 600	1117	77.7	14.8	4027	253
AE14-1	08–15 Dec 2014	37 917	2727	21.7	< D.L.	1499	54.9

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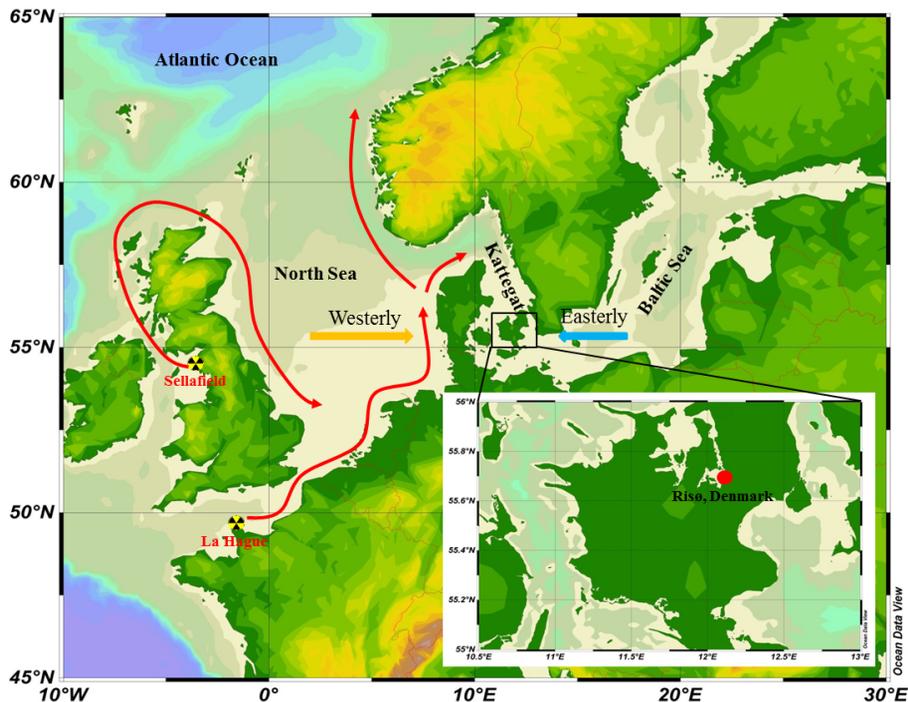
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**Figure 1.** Map showing the sampling site (red dot in the inset) at Risø, Denmark for aerosol collection. The two nuclear reprocessing plants (radioactivity labels), Sellafield (United Kingdom) and La Hague (France) have released a large quantity of  $^{129}\text{I}$  to the marine and atmospheric environment. This anthropogenic  $^{129}\text{I}$  discharged to marine systems has been transported to the North Sea, Baltic Sea, Norwegian Sea, and beyond, by ocean currents (red lines). Yellow and blue arrows show the westerly wind and easterly wind, respectively.

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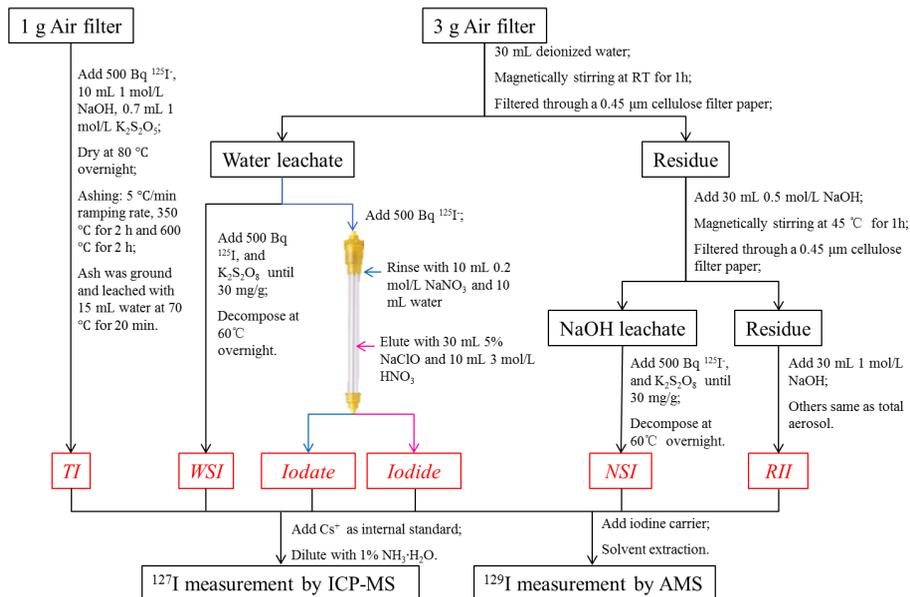
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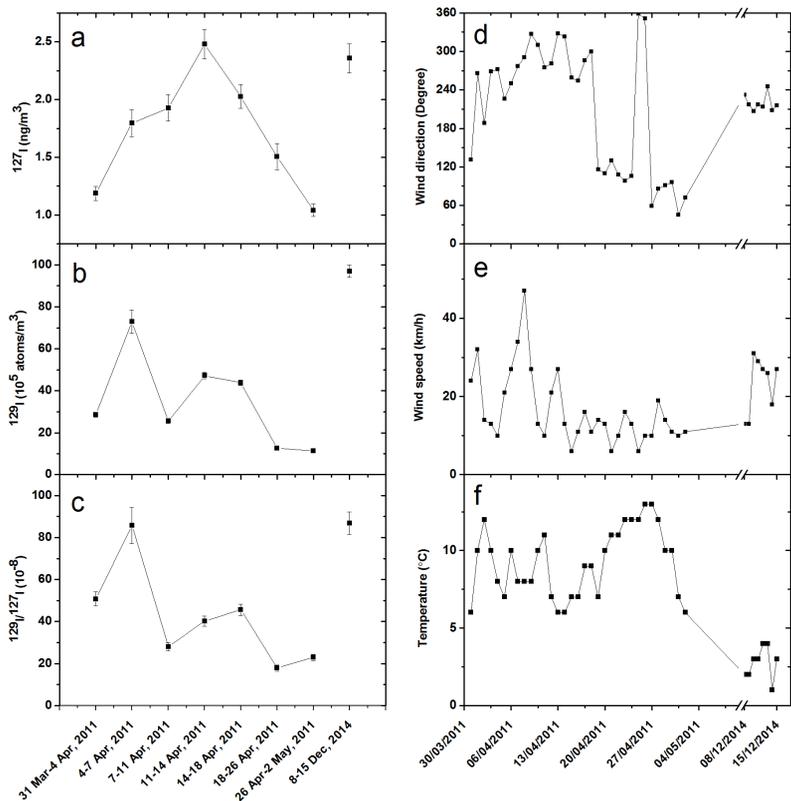
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**Figure 2.** Schematic diagram of the analytical procedure for the determination of  $^{127}\text{I}$  and  $^{129}\text{I}$  species in aerosols. TI for total iodine, WSI for water-soluble iodine, NSI for NaOH-soluble iodine and RII for residual-insoluble iodine.



**Figure 3.** Variation of  $^{127}\text{I}$  concentrations (a),  $^{129}\text{I}$  concentrations (b),  $^{129}\text{I}/^{127}\text{I}$  ratios in aerosols (c), wind direction (d), wind speed (e) and temperature (f) during the sampling period. The historical meteorological data, including temperature, wind direction and wind speed were obtained from the observation station of Hangarvej in Roskilde, Denmark ( $55.594^{\circ}\text{N}$   $12.128^{\circ}\text{E}$ ) based on 41 m a.s.l. (above sea level) (Weather Wunderground webpage, 2015).

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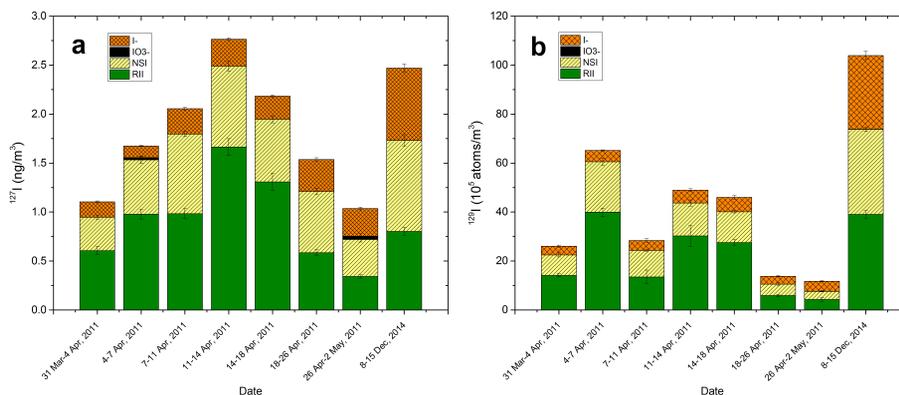
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**Figure 4.** Concentrations of iodine species in the aerosol samples for  $^{127}\text{I}$  (a) and  $^{129}\text{I}$  (b), indicating that NSI and RII are major iodine species and that iodide is the dominant fraction of water-soluble iodine.

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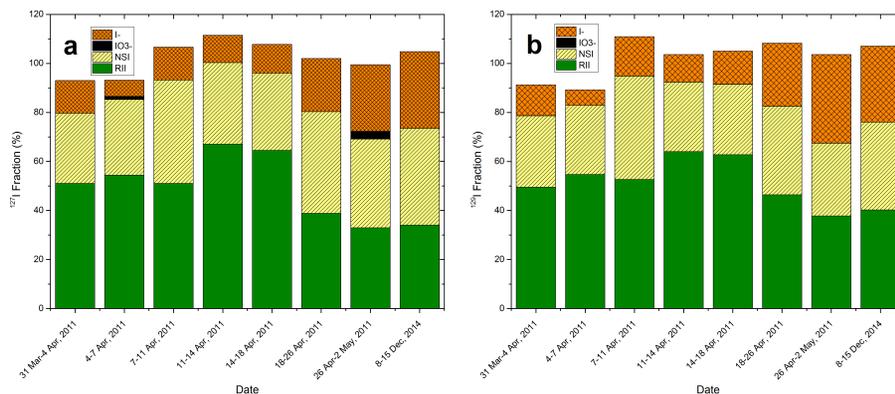
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**Figure 5.** Distribution of iodine species in the aerosol samples for  $^{127}\text{I}$  (a) and  $^{129}\text{I}$  (b), compared with the measured concentrations of total  $^{127}\text{I}$  and  $^{129}\text{I}$  in the original aerosols.

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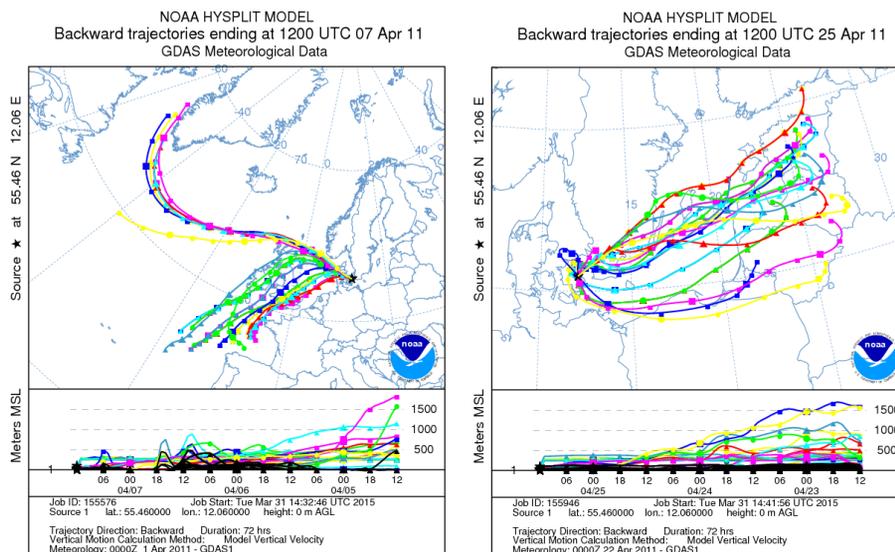
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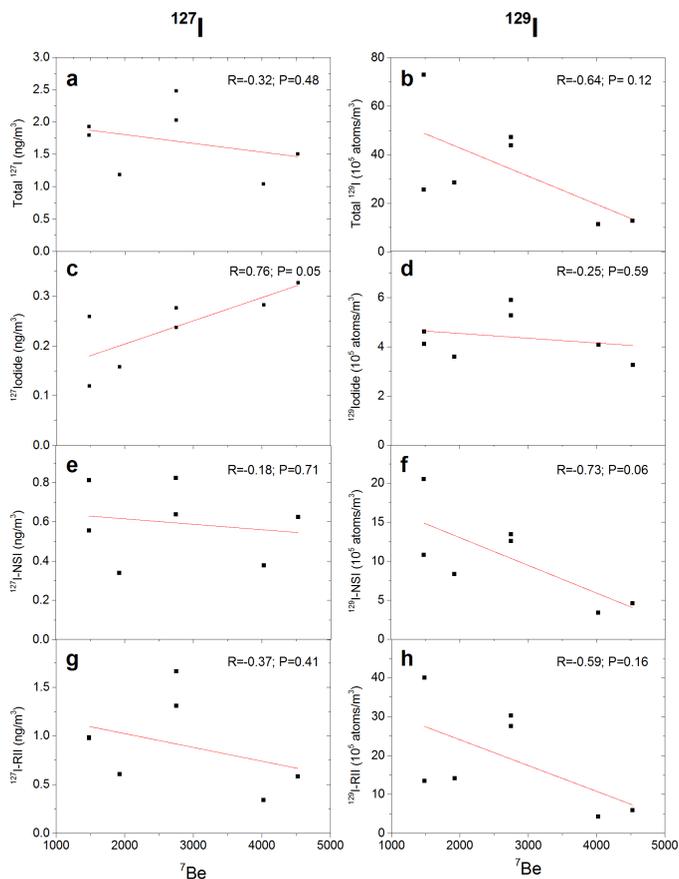
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**Figure 6.** The 72 h (3 days) air mass back trajectories for starting altitudes of 0 m a.g.l. (above ground level) calculated from the FNL database of the National Ocean and Atmospheric Administration (NOAA) and simulated by using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HY-SPLIT) model. 4–7 April 2011 (left) and 21–25 April 2011 (right).

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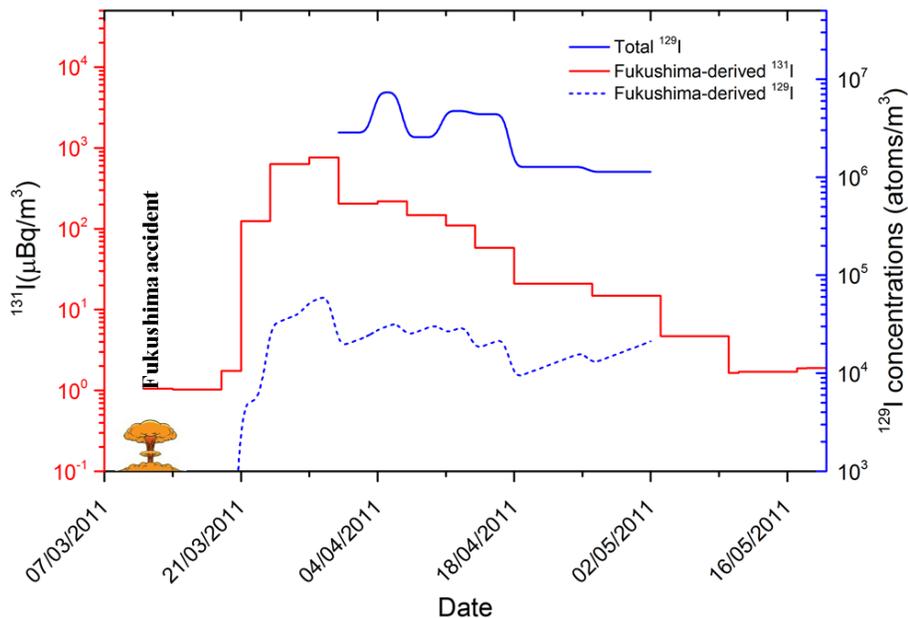


**Figure 7.** Correlation of aerosol  $^7\text{Be}$  with iodine species including total  $^{127}\text{I}$  (a) and  $^{129}\text{I}$  (b),  $^{127}\text{I}^-$  (c),  $^{129}\text{I}^-$  (d), NSI for  $^{127}\text{I}$  (e) and  $^{129}\text{I}$  (f), as well as RII for  $^{127}\text{I}$  (g) and  $^{129}\text{I}$  (h).



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**Figure 8.**  $^{131}\text{I}$  radioactivity (red),  $^{129}\text{I}$  concentrations (blue) in aerosols from Risø, Denmark after the Fukushima accident (Nielsen et al., 2011). The Fukushima-derived  $^{129}\text{I}$  concentrations are calculated based on an  $^{129}\text{I}/^{131}\text{I}$  atomic ratio of  $16.0 \pm 2.2$  deduced from Fukushima-affected aerosol samples (Xu et al., 2015).

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