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Inventories, input and transport of iodine isotopes in Germany

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Introduction and motivation

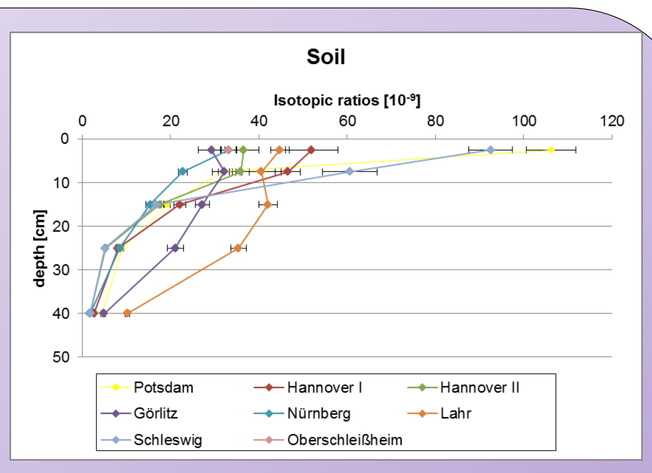
The natural abundances of ^{129}I in the environment have been changed in a sustainable way mainly due to the releases of the European reprocessing plants. The iodine isotopic ratios show a strong disequilibrium. In the North Sea the ^{129}I / ^{127}I ratio has been changed by four orders of magnitude from about $1.5x10^{-12}$ to $3.2x10^{-8}$. Ratios up to 10^{-10} and 10^{-6}, respectively, were found in rain and surface water in Lower Saxony [1].
Iodine from precipitation is accumulated in soils, infiltrates groundwater, is transported by surface water and makes its way through the biosphere. Still, most of the ecological pathways of iodine are unclear.
The goal of this project is to analyse the inventories in the pedosphere, study the continuous input from the atmosphere via wet and dry deposition and the output per river water of ^{129}I, ^{127}I and the isotopic ratio ^{129}I / ^{127}I resp.

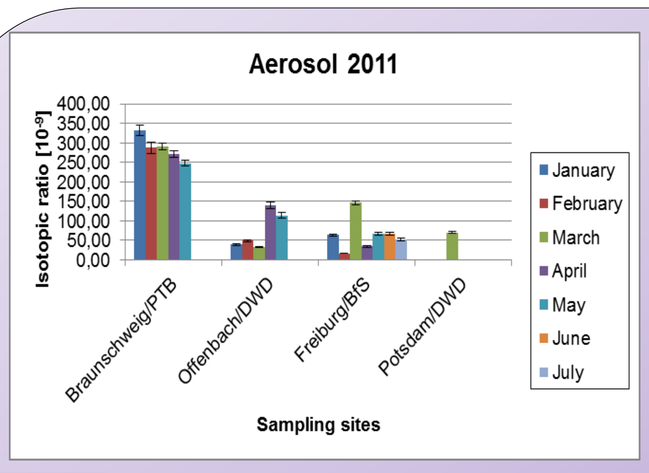
Methodology

Iodine was extracted from the soils by dry combustion with excess oxygen and nitrogen. Separation of iodine from water samples was carried out using ion exchange resin. The aerosol filters were subjected to an alkaline leaching, the iodine in the resulting solution was separated from the matrix by liquid-liquid exchange using chloroform. The concentration of the stable ^{127}I was measured with ICP-MS (inductively coupled plasma – mass spectrometry) and the concentration of ^{129}I and the $^{129}I/^{127}I$ ratio was determined by Tandy AMS system (accelerated mass spectrometry) at the ETH Zurich. To select the charge state 2+ a terminal voltage of 0.5 MV was used [2,3].

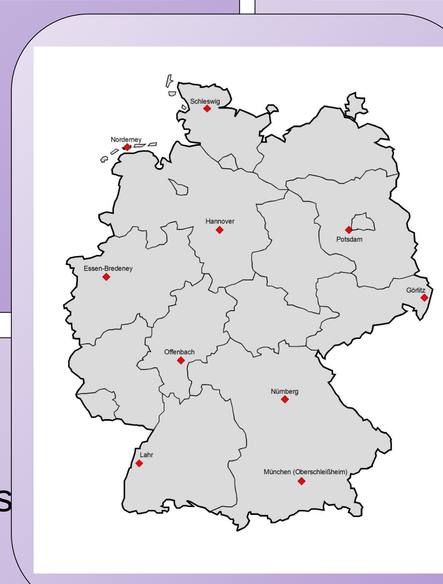
Results

The ^{129}I inventories of the studied soil profiles (0-50 cm depth) ranged from 120 mBq m^{-2} to 320 mBq m^{-2}. In most cases, the $^{129}I/^{127}I$ isotopic ratios of the top layers (0-5 cm, 5-10 cm) were several magnitudes higher than those of the deeper layers. This suggests ^{129}I downward migration being a very slow process.
First results indicate that ^{129}I inventories of soils are slightly higher in the northern and western parts of Germany than in the southern and eastern parts.

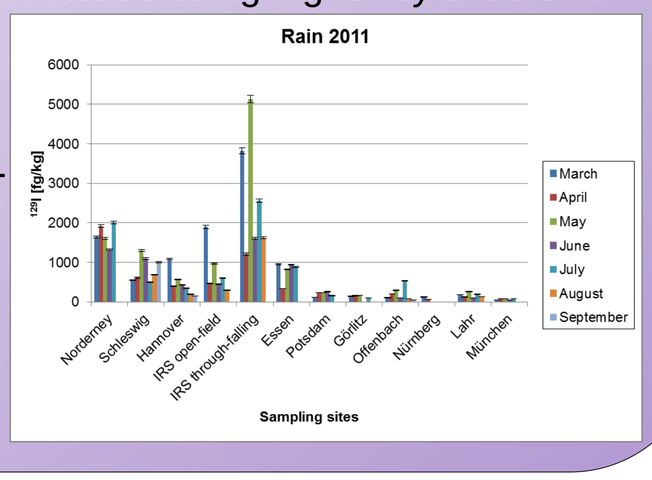




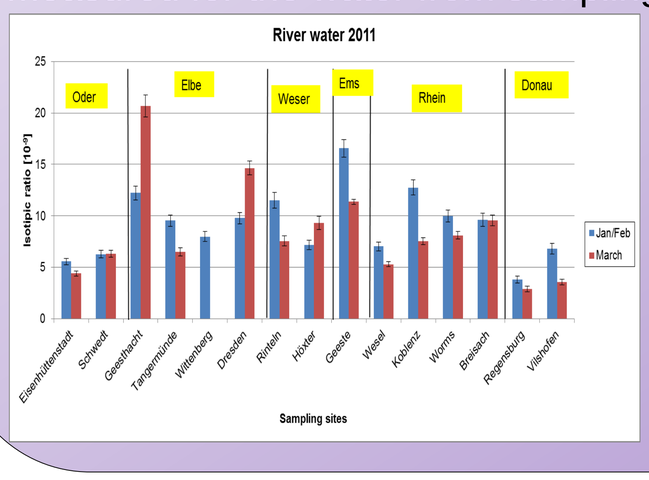
Dry deposition is determined by analyses of aerosol filters, which collect the particle-bound iodine from the athmosphere.
The figure exhibits temporal variations of the $^{129}I/^{127}I$ isotopic ratios at four different sampling sites. They range from $0.2x10^{-8}$ to 30 $x10^{-8}$, with Braunschweig (northern Germany) showing the highest values.



Concentrations of ^{129}I in rain water decrease from north to south, and from west to east, respectively. They range from 48 - 1,093 fg L^{-1}. The isotopic ratios range from $2.9x10^{-8}$ to $4.9x10^{-7}$.
A comparison of open-field and through-falling precipitation, resp., revealed ^{129}I values being higher by a factor of about 4 for the through-falling rain.
This is probably because it contains the dry deposition from the leaves.



River water is collected at 15 sampling sites along the major rivers in Germany. The isotopic ratios of the river water range from $2.9x10^{-9}$ to $20.7x10^{-9}$. The highest values were measured in the north (Geeste/ Ems, Geesthacht/Elbe) and the lowest values were measured for the water from sampling sites in the southeast of Germany at the River Donau and in the east at the Oder River. However, differences among the values are small.



Conclusion

The results of the soil samples are as expected, with the highest ^{129}I content in the topsoil. For both aerosol-samples and water (precipitation and river water) we found higher values for the ^{129}I content and the isotopic ratios, resp., at the sampling sites in the north and west than in the south and east of Germany.
This is most likely due to the fact that man-made ^{129}I from the reprocessing plants is dispersed from the North Sea to the continent by atmospheric transport. Further sampling and measurements are required to validate this theory.

References

[1] Michel et al. (2012) Science of Tot Enviroment **419** 151-169. [2] Tosch (2010), Dissertation, Leibniz Universität Hannover, http://www.zsr.uni-hannover.de/ arbeiten/ drtosch.pdf [3] Daraoui et al. (2011) *submitted to J. Environ. Rad.* [4] Szidat et al. (2000) *Nucl. Instr. Meth. Phy. Res. B* **172**, 699-710. [5] Ernst et al. (2003) *Kerntechnik* **68** (4), 155-167.

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