Project acronym: **Euro-limpacs**

Project full name: **Integrated Project to evaluate the Impacts of Global Change on European Freshwater Ecosystems**

Instrument type: **Integrated Project**

Priority name: **Sustainable Development**

**Deliverable No. 25**

**Summary of 9 months of experimental manipulations at Gårdsjon catchments**

Due date of deliverable: 31 January 2005
Actual submission date: **30 July 2006**

Start date of project: **1 February 2004**  
Duration: **5 Years**

Organisation name of lead contractor for this deliverable: **9. IVL**

The majority of this report was prepared by partner **9. IVL**.

Revision [1.0]

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**Project co-funded by the European Commission within the Sixth Framework Programme (2002-2006)**

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<tr>
<th>Dissemination Level (tick appropriate box)</th>
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<tbody>
<tr>
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<td>PP</td>
<td>Restricted to other programme participants (including the Commission Services)</td>
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<td>RE</td>
<td>Restricted to a group specified by the consortium (including the Commission Services)</td>
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<tr>
<td>CO</td>
<td>Confidential, only for members of the consortium (including the Commission Services)</td>
</tr>
</tbody>
</table>
1 Irrigation experiment in Gårdsjön Catchment, Sweden

1.1 Objectives and hypothesis

Future climate scenarios suggest large variabilities in precipitation amounts in different parts of Europe. In some regions, large increases are expected. Increased precipitation will potentially affect the mobility of mercury and other pollutants accumulated in forest soils and can thus lead to increased loadings on surface waters. The objective of this activity in WP 5 is:

- To determine the influence of increased precipitation on the release of total mercury and methylmercury from forest soils to surface waters.

The main hypothesis is:

Increased precipitation will:

- Change the hydrological pathways in the soil and enhance the leaching of total mercury and methylmercury
- Lead to the formation of anaerobic zones in the soil which will enhance formation of methylmercury

1.2 Experimental design and sampling strategy

1.2.1 Site description

The research site at Gårdsjön is located at 130 m elevation, 12 km inland on the Swedish west coast (58º 04' N, 12º 03' E), Figure 1. The 6300 m² catchment G1 has a 85 to 105 years old Norway spruce (Picea abies (L.) Karst.) forest, with some Scots Pine (Pinus sylvestris) present. The bedrock is gneissic with intermediate to acid chemical composition. The soils are podsols. The average soil profile consisted of a ca 10 cm thick humus layer (LFH). This was underlain by a ca 7 cm thick layer of eluviated mineral soil (A) with a distinct light grey colour. This, in turn, was underlain by 17 cm of lower mineral soil (B) and then a further 14 cm of the lowermost mineral soil (BC), which is a transition between B and unaltered glacial till (C). A detailed description of the experimental area is given by Andersson and Olsson (1985) and by Andersson et al., (1998).
Catchment G1 was in 1991 fitted with a 7000 m$^2$ plastic roof and treated with an artificial clean precipitation sprinkled underneath the roof between 1991 and 2001 in a large de-acidification experiment (Hultberg and Skeffington, 1998). The plastic sheeting was removed in summer 2001 and most of the activities were suspended. The monitoring of runoff chemistry and volume was however maintained. Re-opening of the G1 catchment brought much increased level of activities. There was a need to restore the watering system by installing new sprinklers and to remove the old system. It turned out necessary to remove and partly to replace the old wooden walkways, which practically rotted away. The site was also swept for old and unused installations of various collectors, samplers and other kinds of scientific litter. The downside of this effort was that it meant much increased presence of people in the catchment and therefore an increased risk of damaging the soil by walking on it. This is a serious factor that needs to be taken into account when designing the field activities and when analysing results. A view from under the roof in of catchment G1 is presented in Figure 2.
The lower part of NW slope of the catchment is relatively steep and inaccessible. Therefore the density of various samplers, other installations and of human traffic and thus a potential damage to the soil was less there comparing to flatter parts of the catchment. That part of the catchment (ca 1000 m²) was chosen for watering campaign July 2004. Using only a part of the catchment - about one sixth of the total area of 6300 m² - requires less water. That opened for a possibility of longer watering, which is normally limited by the capacity to produced de-ionised water. In the 2005 experiments, the complete catchment area was irrigated.

1.2.2 Irrigation system and sampling facilities

Sprinkling system was restored by replacing most of the tubing and all sprinklers in July 12 - 16th. The capacity was increased by installing 20 mm distribution pipes instead of 16mm and by installing 242 new sprinklers (see Figure 3) with capacity of 80 l/hour/sprinkler (3.1 mm/h for 6300 m² G1 catchment).
The runoff at the G1 ROOF catchment was sampled automatically at a frequency proportional to the flow. The samples were routinely analysed for a total of 10 major and trace elements. Aluminium is speciated into organic (Alo) and inorganic (Ali) fractions using a method adapted from Driscoll (1984). Samples were also analysed for DOC and for total Hg and MeHg.

1.2.3 Additions of stable isotopes

Before the irrigation experiment started, four 1m² sites, three inside G1 catchment and one outside G1 catchment, were selected for addition of ¹⁹⁸Hg isotope. Soil cores were collected before, during and after the irrigation experiment. By analysing the fraction of ¹⁹⁸Hg which had been converted to methylmercury, a measure of the methylation rate in the soil can be obtained.

MMHg in the soil samples was analysed by N₂-distillation, NaBE₄ ethylation and GC-CVAFS. THg were analysed by H₂SO₄-HNO₃ digestion, BrCl oxidation, SnCl₂ reduction and CVAFS. TOC in the soil samples was analysed by loss on ignition. The mercury isotope composition in the MMHg and THg was analysed by ICP-MS.

2 Preliminary results

The total amount of irrigation in the 2005 experiment is presented in Figure 4. The added amount corresponds roughly to 2 -3 times the monthly natural precipitation (throughfall) in the months May, June, July and September, and an equal amount in October. No irrigation was performed in August.
In Figure 5, preliminary results of total and methylmercury concentrations in run-off are presented. Initially during the irrigation period, more or less stable concentrations total and methylmercury in runoff were observed. The artificial irrigation caused an increased runoff and the flux of mercury from the catchment increased correspondingly, in addition to the moderate concentration increase. After about 3.5 months (August 2005), a sharp increase in methylmercury concentration occurred. The concentrations of methylmercury increased from the normal level below 0.1 ng/L to averages around 1 ng/L and individual values above 5 ng/L. Concurrent increases in DOC, and indications of sulphide in runoff water, suggested that the soil was saturated with water and that anaerobic conditions in the soils had been established i.e. favouring methylation of mercury via sulphate reducing microorganisms. The irrigation was stopped in October 2005 and methylmercury concentrations decreased somewhat but remain above "normal" levels (i.e. 0.1 to 1 ng/L in comparison to <0.1) until present date (December 31, 2005).
In figures 6 and 7, the annual fluxes of total- and methylmercury, respectively, are presented. The increase in flux of total mercury is more or less proportional to the increase in runoff, caused by the irrigation. The increase in methylmercury flux is significantly larger indicating that other factors than hydrology has influenced the concentration in runoff water.

**Figure 6.** Annual fluxes of total mercury from irrigated catchment G1. The red bar indicates the year when the experimental irrigation was performed.
2.1 Preliminary conclusions

- First irrigation phase (May-June) with about 60% additional precipitation, did not affect TotHg or MeHg concentrations. Fluxes increase equivalent to change in run-off amounts.
- After warm and wet summer, large increases in MeHg, TotHg, DOC (even before second irrigation phase). Also reduced sulphur.
- High levels of MeHg, TotHg and DOC continued during second irrigation period (and after).
- Increased wetness of soil led to anaerobic conditions, SO4 reduction, methylation. Also increased DOC, TOC.

3 References

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