ON THE SOURCES OF PCBs AND PBDEs TO LAKE MJØSA, NORWAY

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Introduction

Lake Mjøsa is the largest lake in Norway, with a surface area of 365 km^2. High levels of some contaminants in fish from the lake[1,2] have caused public attention and concerns, and health authorities recommend that the consumption of certain species should be restricted. This study describes key results from an initial study aimed at identifying and quantifying the sources that control current levels of selected contaminants in lake Mjøsa[3]. An important objective has been to evaluate the relative significance of the atmospheric pathway, including an assessment of local versus long-range atmospheric transport for selected PCBs (polychlorinated biphenyls) and PBDEs (polybrominated diphenyl ethers). A multimedia fate model has additionally been developed and evaluated as part of the project to evaluate (*i*) if the levels of PCBs in the lake can be explained by the current understanding of existing sources, as well as (*ii*) if there are significant knowledge gaps related to the sources, pathways and environmental fate of PCBs in lake Mjøsa.

Materials and Methods

Sampling and analysis

A limited sampling campaign was carried out during the autumn of 2004 and beginning of 2005 to measure selected PCBs and PBDEs in air, lake water and river water (Figure 1). Additional samples from sewage treatment plants (STPs) were collected and analysed within another project to evaluate discharge of contaminants into lake Mjøsa[4].

In brief, 22 bulk air samples were collected using a conventional high volume air sampler[5] that were deployed at Kise to evaluate temporal patterns in air. Each air sample was collected over a time period of 48 hours. In addition, 10 passive air samplers using polyurethane foam as sampling medium[6,7] were deployed to evaluate spatial patterns in air and harvested after a sampling period of 12 weeks. Furthermore, 4 water samples (>257 L) were collected from the lake (close to Kise) in September 2004, and additional passive water samples were collected from the lake using SPMDs (semipermeable membrane devices)[8] as sampling medium.

All samples were analysed at NILU. The samples were spiked with internal standards (^{13}C-PCBs and PBDEs) prior to the sample preparation. Both filters and polyurethane foams were Soxhlet extracted with n-hexane/diethylether (9/1). The extracts were concentrated, treated with concentrated sulphuric acid, and further cleaned on a 4 g silica column with n-hexane/diethyl ether (9/1). Both PCBs and PBDEs were analyzed with GC/HRMS in two separate runs.

Model description

A fugacity-based[9] multimedia fate model was furthermore developed and parameterised for lake Mjøsa. The model is similar in structure to the QWASI (Quantitative Water Air Sediment Interaction) model by Mackay et al.[10], except that this model was made non-steady state. The processes included and process descriptions are identical to the Oslofjord POP model[11], except that spatial resolution in this case was ignored. A detailed account of all process descriptions and input parameters is given elsewhere[3,11].

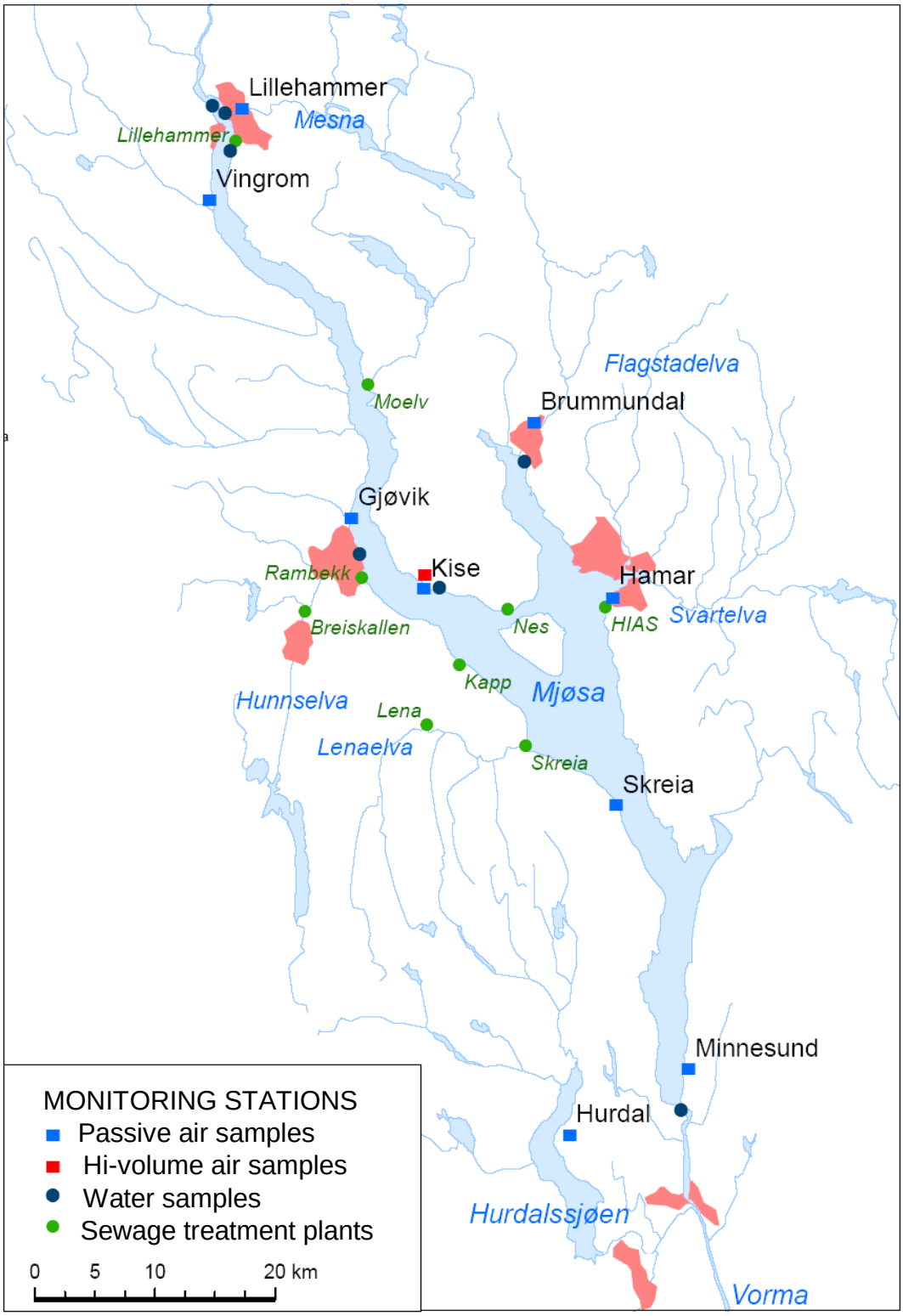

Figure 1. Monitoring stations around lake Mjøsa (map by the Norwegian Pollution Control Authority).

Results and Discussion

PCBs

In general, the atmospheric burden of PCBs around the lake was found to be fairly typical for remote levels in Norway. Elevated levels of PCBs in air as measured in the vicinity of the lake (Kise), was found to coincide with transport episodes from potential source regions outside the lake region, using backward air trajectories from the FLEXTRA model[12,13]. The Oslo area, Great Britain and Eastern Europe / Russia are suggested as likely source regions for the elevated levels that were recorded during the monitoring campaign that was carried out.

The passive air samplers were used to assess potential local source regions. The atmospheric levels were found to be slightly higher in two of the larger towns around lake Mjøsa (Hamar and Gjøvik) as compared to the other stations around the lake. In comparison, levels of PCBs in Oslo were found to be ten to fifty times higher as compared to the typical atmospheric burden around lake Mjøsa, thus confirming that large urban centres tend to be regional "hot spots" of PCB emissions[7].

The model that was developed and parameterised for the lake underestimates observed levels of PCBs by a factor of ~4, whilst the congeneric patterns was satisfactorily reproduced (in lake water and sediments). Deviations between predicted and observed concentrations are attributed to two possible causes: (*i*) the true fluxes of PCBs to the lake may be underestimated (*ii*) an underestimate of PCB loss processes in the model. At present, it is not possible to assess the relative importance of these two errors, based on the current empirical knowledge.

Overall, the results suggest a fairly complex source contribution of PCBs to lake Mjøsa, and that significant fluxes of PCBs are yet to be identified. The current understanding of terrestrial sources of PCBs to the lake is limited and probably underestimated. Occasional floods and heavy rainfalls may cause pulses of untreated discharges from the STPs (bypass), and are expected to cause pulses of PCBs into the lake. Particle-mediated transport of PCBs along river bottoms is an additional source that is yet to be accurately described. The results of this project furthermore indicate that the atmosphere is a key source of PCBs to lake Mjøsa, in comparison to initial estimates of the amounts discharged by rivers as well as sewage treatment plants. Further efforts to reduce primary atmospheric emissions of PCBs (nationally and internationally) may therefore in the long run contribute to reduce the environmental levels in lake Mjøsa. Before reliable source reduction measures can be determined, an improved characterisation of the non-atmospheric inputs (i.e. local discharges) is strongly recommended. Specifically, an improved quantitative understanding of the relative significance of current sources on land versus sources in the lake (e.g. leaching of PCBs from surface sediments contaminated in the past) is urgently needed, as sediments nevertheless constitute an important reservoir of PCBs. It is recommended that possible follow-up studies should include further investigations that seek to improve the quantitative understanding of exchange processes occurring between water and sediments, in order to obtain a better grip on the relative importance of current sources and sediment leaching (the latter due to contamination in the past).

PBDEs

In comparison to the PCBs, PBDEs exhibit a greater spatial and temporal variability in air. The town of Lillehammer is identified as a local atmospheric source region for key congeners that were included in the technical mixture "penta-BDE", experiencing even higher levels than the city of Oslo. The long-term atmospheric burden as derived from passive samplers still suggests a limited potential for atmospheric distribution from Lillehammer to other areas around the lake. Based on hi-volume air measurements at Kise, it is found that that $\Sigma PBDE_5$ (BDE-28, 47, 99, 100 and 153) exhibit a significant temporal variability. A trajectory-based analysis of samples experiencing elevated levels of $\Sigma PBDE_5$, indicate both episodes of local transport (presumably from Lillehammer) as well as long-range atmospheric transport (possibly from Great Britain). An interesting observation is that the average atmospheric concentrations of $\Sigma PBDE_5$ is now equal to BDE-209, even though BDE-209 has been assumed to have a fairly limited potential for atmospheric transport in comparison to other PBDE congeners[14]. The results for BDE-209 finally indicate that Great Britain may occasionally be a source region for BDE-209 to the atmospheric environment of lake Mjøsa.

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