Sources of Environmental Contaminants into Lake Mjøsa

Action-oriented prestudy

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Authors:	Knut Breivik, Martin Schlabach, Torunn Berg

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NILU

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Preface

The purpose of this action-oriented prestudy has been to gain insight on the sources that control contemporary levels of selected environmental contaminants in Lake Mjøsa. The study was led by the Norwegian Institute for Air Research (NILU), with the help of Eirik Fjeld and Gösta Kjellberg of the Norwegian Institute for Water Research (NIVA).

We would like to thank the Norwegian Pollution Control Authority (SFT) for funding this project, and the Norwegian Research Council for funding supplementary PBDE measurements. We would also like to thank the people who have contributed useful information to the project work. This particularly applies to Eirik Fjeld and Gösta Kjellberg at NIVA, who have been of invaluable help in connection with sample collection during this project and data collection from previous studies.

We would also like to thank Elin Lundstad (Norwegian Meteorological Institute) for meteorological data for Kise, employees of the Norwegian Centre for Soil and Environmental Research (Jordforsk) for samples from treatment plants along Lake Mjøsa, and Sverre Solberg (NILU) for trajectory calculations. We would also like to thank the people who have assisted with the sample collection, and the employees who were involved with this work at the Norwegian Crop Research Institute in Kise.

I would like to thank Jon L. Fuglestad, who was responsible for this study at the Norwegian Pollution Control Authority.

Kjeller, 14 September 2005.

Martin Schlabach
Project Manager, NILU

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1. Extended abstract in English

Lake Mjøsa is the largest lake in Norway, with a surface area of 365 km^2. High levels of some contaminants in fish have caused public attention and concerns, and health authorities recommend that the consumption of certain species should be restricted. This report describes key results from an initial survey aimed at identifying and quantifying the sources that control levels of selected contaminants in lake Mjøsa. 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), PBDEs (polybrominated diphenyl ethers) as well as Hg (mercury).

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. It is emphasised that the empirical data presented and discussed are still limited. Further research and complementary surveys are therefore recommended to improve current knowledge in order to provide decision makers with a sound basis for further development of sensible control strategies.

PCBs

Overall, the results suggest a fairly complex source contribution of PCBs to lake Mjøsa, and that significant fluxes of PCBs may yet to be identified. The results of this project furthermore suggest 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 (STPs).

The atmospheric burden of PCBs around the lake is fairly typical for remote levels in Norway. Elevated levels of PCBs in air as measured in the vicinity of the lake (Kise), coincide with transport episodes from potential source regions outside the lake region. The Oslo area, Great Britain and Eastern Europe / Russia are suggested to be key contributors to the elevated levels that were recorded during the monitoring campaign that was carried out between September 2004 and January 2005. A campaign was also carried out deploying a limited number of passive air samplers to evaluate spatial patterns in air in order to assess potential local source regions. The atmospheric levels were slightly higher in two of the larger towns (Hamar and Gjøvik) as compared to the other stations around the lake. In comparison, the levels of PCBs in Oslo were found to be ten to fifty times higher as compared to the typical atmospheric burden around lake Mjøsa.

The model that has been developed to derive source-receptor relationships for the lake clearly underestimates observed levels of PCBs, whilst the congeneric patterns seem to be satisfactorily reproduced (in lake water and sediments). The results indicate a complex picture whereby the environmental levels in the lake can be explained by both historical and current inputs. Deviations between predicted and observed concentrations are attributed to two key causes: (*i*) the true fluxes of PCBs to the lake may be underestimated (*ii*) errors in the model (e.g. an underestimate of the relative importance of sediment leaching). Regrettably, it is virtually impossible to assess the relative importance of the two, based on the current empirical knowledge. Still, the overall results indicate that the current understanding of landbased sources of PCBs to the lake is limited and probably underestimated. Occasional floods and heavy rainfalls may cause pulses of untreated discharges bypassing the STPs, and are expected to cause elevated inputs of PCBs to the lake during such events. Particle-

mediated transport of PCBs along river bottoms is an additional source that is yet to be accurately described.

The results provide useful information for identifying key knowledge gaps, but uncertainties in existing knowledge currently mitigate the potential for identifying reliable and sensible control strategies. The atmosphere is identified as a key pathway of PCBs to the lake, and elevated levels coincide with transport events from urban areas in Norway (Oslo) and from abroad. Further efforts to reduce primary atmospheric emissions of PCBs (nationally and internationally) may therefore contribute to reduce the environmental levels and exposure in vulnerable ecosystems, such as Mjøsa. Before reliable source reduction measures can be prioritised and assessed locally, an improved characterisation of the non-atmospheric inputs (i.e. local discharges) is strongly recommended. Specifically, an improved understanding of the relative significance of current sources on land versus sources in the lake (e.g. leaching from surface sediments contaminated in the past) is urgently needed. Sediments neverthelss constitute an important reservoir for these compounds. 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). Finally, it cannot be excluded that contaminated material containing PCBs may have been dumped in the lake in the past, causing a significant contribution to current environmental levels. Potential investigations should anyhow address the lake as a whole as restrictions on fish consumption are in effect for the entire lake ecosystem.

PBDEs

In comparison to the PCBs, the PBDEs exhibit a greater spatial and temporal variability. 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 centre of Oslo. The long-term atmospheric burden as derived from passive samplers still generally 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 (presumably 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 is assumed to have a fairly limited potential for atmospheric transport in comparison to other PBDE congeners. 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.

Hg

Atmospheric deposition of mercury to Lake Mjøsa has been studied on basis of precipitation samples collected during September 2004-May 2005 and on basis of previous studies. The results indicate that atmospheric long-range transport is the major source. The input to Lake Mjøsa from this source has been estimated to be 2 kg. However, sewage treatment plants (STPs) seems also to be a significant source of mercury and an indicative number of the input to Mjøsa from these could be about 0.15-3 kg. National emission data in addition to data from moss surveys indicate that emission sources of mercury to air seems to be of limited significance in this region.

2. Extended abstract in Norwegian

Høye nivåer av enkelte miljøgifter i Mjøsa har skapt bekymring, og kostholdsråd har blitt innført for å minimere risiko ved konsum av enkelte fiskearter. Denne rapporten beskriver sentrale resultater fra en tiltaksorientert forundersøkelse med formål å belyse hvilke tilførsler som kontrollerer nivåer av utvalgte miljøgifter i Mjøsa. Fokus har særlig vært rettet mot betydningen av atmosfæriske tilførsler, og en evaluering av lokal kontra atmosfærisk langtransport for polyklorerte bifenyler (PCB), polybromerte difenyletere PBDE (BDE-209) og kvikksølv (Hg). En datamodell har også blitt utviklet og evaluert i prosjektet for å undersøke (*i*) om nivåene av PCB i innsjøen kan forklares ut fra dagens kunnskap om eksisterende tilførsler, samt (*ii*) om det er sentrale kunnskapsmangler vedrørende tilførsler og oppførsel av PCB i Mjøsa. Det presiseres at datagrunnlaget for de vurderinger og beregninger som har blitt foretatt gjennomgående er begrenset, og at oppfølgende undersøkelser bør foretas for å bedre kunnskapsgrunnlaget og dermed også beslutningsgrunnlaget.

PCB

Samlet vurdert tilsier resultatene at tilførselsmønsteret for PCB til Mjøsa er sammensatt, og at det kan være betydelige tilførsler av PCB som ennå ikke er identifisert. Resultatene fra dette prosjektet tyder imidlertid på at atmosfæren er en viktig kilde av PCB til Mjøsa, sammenlignet med en første beregning av tilførsler via elver (basert på beregnede konsentrasjon i vann) samt renseanlegg (under normale operasjonsforhold). Nivået av PCB i luft rundt Mjøsa er imidlertid nokså typisk for bakgrunnsbelastningen i Norge, mens episoder med forhøyet belastning primært skyldes langtransport fra områder utenfor Mjøsregionen (Osloregionen og utlandet). Videre tiltak for å redusere utslipp av PCB til luft bør derfor vurderes, sett i sammenheng med nasjonale planer for utfasing av PCB og gjennom videre oppfølging av internasjonale avtaler.

Modellen som har blitt utviklet for å belyse sammenhenger mellom tilførsler og nivåer i innsjøen underestimerer observerte konsentrasjoner av PCB. Resultatene viser et sammensatt bilde der nivåene av PCB i Mjøsa kan forklares ut fra eksisterende og historiske tilførsler. Modellen som tar hensyn til både eksisterende og historiske tilførsler, kan forklare om lag en fjerdedel av de observerte nivåer av ΣPCB_7, basert på vannprøver tatt utenfor Kise. Avviket mellom observerte konsentrasjoner forventes primært å kunne skyldes to forhold; (*i*) Underestimering av tilførsler av PCB til innsjøen. (*ii*) Feil og mangler ved modellen. På grunn sistenevnte forhold (usikkerheter i modellen) kan man derfor ikke anta at de reelle tilførsler er fire ganger høyere enn den beregnede tilførsel på 0.2 kilo ΣPCB_7 for 2004. Resultatene tilsier imidlertid at landbaserte tilførsler av PCB kan være underestimert. Tiltak lokalt bør derfor i første omgang fokusere på å oppnå en forbedret karakterisering av lokale landbaserte tilførsler. Det kan heller ikke utelukkes at kilder i innsjøen (*utlekking av PCB fra belastede sediment, eventuelt PCB-holdig avfall som kan ha havnet i innsjøen*) kan gi et signifikant bidrag til det generelle belastningsnivået i Mjøsa. Sedimentene utgjør uansett et viktig reservoar for PCB. Videre undersøkelser bør derfor forsøke å bedre systemforståelsen for de prosesser som er bestemmende for utveksling av PCB mellom vann og sediment i Mjøsa for å oppnå et bedre estimate på tilførsler som skyldes tidligere tiders utslipp.

PBDE

Atmosfæriske nivåer av PBDE utviser en betydelig variasjon over tid og rundt Mjøsa. Lillehammer utpeker seg som kildeområde til luft rundt Mjøsa for sentrale enkeltforbindelser som inngår i den tekniske blandingen penta-BDE. Nivåene i luft av fire utvalgte enkeltforbindelser ($\Sigma PBDE_4$ = BDE-28, 47, 99, 100) var over en periode over flere måneder høsten 2004 ca. 14 ganger høyere enn gjennomsnittet for de andre stasjonene rundt Mjøsa. Kilden(e) til utslippet av $\Sigma PBDE_4$ i Lillehammer kan imidlertid ikke identifiseres på bakgrunn av eksisterende data, og man vet heller ikke hvilken innvirkning den forhøyede atmosfæriske belastningen har på nivåer av PBDE i innsjøen. De forhøyede nivåer av $\Sigma PBDE_4$ synes over tid å være begrenset til Lillehammer-regionen, noe som tilsier at utslippene i Lillehammerregionen *generelt* har et begrenset potensiale for spredning til sørlige deler av Mjøsa. Basert på luftmålinger på Kise av $\Sigma PBDE_5$ (BDE-28, 47, 99, 100 og 153), så synes $\Sigma PBDE_5$ i større grad enn PCB å være kontrollert av transportepisoder med forhøyede nivåer. Resultatene tilsier at episodene for $\Sigma PBDE_5$ på Kise skyldes både lokal atmosfærisk transport fra Lillehammer-regionen og langtransport (Storbritannia), mens forhøyede nivåer av BDE-209 (deka-BDE) primært ser ut til å skyldes langtransport (Storbritannia). Resultatene fra Kise dokumenterer forøvrig at det generelle atmosfæriske belastningsnivået for BDE-209 ved Kise nå er på samme nivå som $\Sigma PBDE_5$.

Kvikksølv

Prosjektet har vurdert atmosfærsike tilførsler av kvikksølv (Hg) til Mjøsregionen på basis av innsamlede nedbørsprøver fra september 2004 til begynnelsen av juni 2005 og resultater fra tidligere undersøkelser. Resultatene tilsier at tilførsler av Hg fra atmosfærisk langtransport er største kilde. Det kan imidlertid ikke utelukkes at det er lokale kilder som påvirker luftkvaliteten lokalt. Tilførselen av kvikksølv til Mjøsa gjennom nedbør er estimert til å være 2 kg. Et indikativt estimat for kvikksølv fra renseanlegg er beregnet til å være i området 0.15-3 kg. Det er behov for ytterligere og mer omfattende undersøkelser for å forbedre de foreløpige estimat for tilførsler av kvikksølv til Mjøsa.

For å få et bedre beregninger til bruk i modeller anbefales det å måle totalkvikksølv og metylkvikksølv i nedbør over minimum et år. For å kunne studere atmosfærisk langtransport bedre anbefales det å måle med kvikksølvmonitor for å få bedre oppløsning på dataene. Det store usikkerhetsnivået på prøver fra renseanlegg tilsier at det må tas mange prøver og at både prøvetaking og analyser må kvalitetssikres bedre.

3. Background and Objectives

Lake Mjøsa is the largest lake in Norway, with a surface area of 365 km^2. Some 200,000 people live within Lake Mjøsa's drainage basin, and about 150,000 of these people live in the cities of Hamar, Lillehammer and Gjøvik, and in towns of varying size. Past pollution of Lake Mjøsa in the 1960s and 1970s led to major changes to the lake, including frequent flourishing of algae. However, measures that were part of the "Lake Mjøsa action plan" in the 1980s and 1990s have yielded results, and led to a substantial improvement to water quality. During the last few years, awareness has increased of a number of problem compounds (environmental contaminants), creating new cause for concern (Fjeld et al. 2004). Dietary advice has been introduced to minimize the risk associated with consuming certain species of fish. However, an evaluation of sensible control strategies to reduce the burden on the environment requires an understanding of quantitative relationships between sources and levels of contaminants in Lake Mjøsa (source–receptor relationship). This report describes the key results of an action-oriented prestudy to reveal the sources that control levels of selected contaminants in Lake Mjøsa. The project focused on three of these environmental contaminants: polychlorinated biphenyls (PCB), polybrominated diphenyl ethers PBDE (BDE-209) and mercury (Hg). The structural formulas of PCBs and PBDEs are illustrated in Figures 1 and 2.

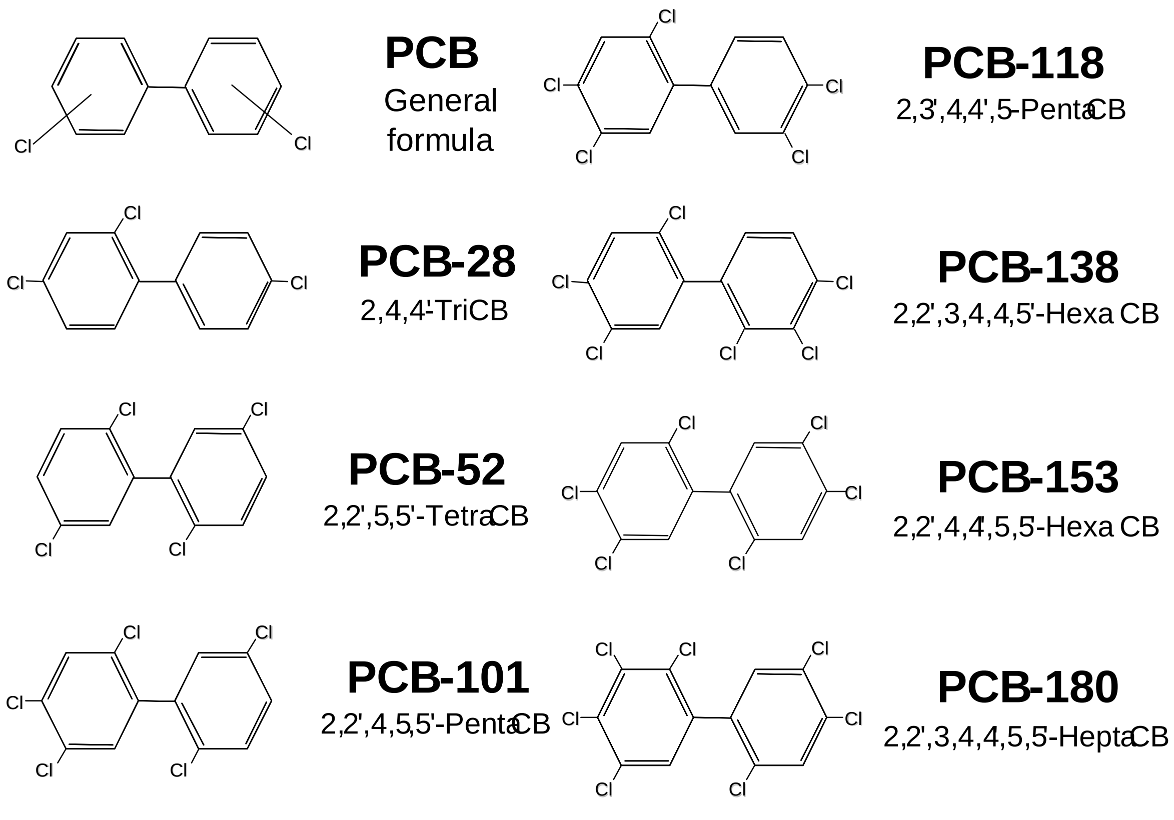

Figure 1. Structural formula for PCB and seven key PCB compounds (congeners) that are discussed in this report.

While mercury is an element with both anthropogenic and natural emission sources, PCBs and PBDEs are industrial organic chemicals that have been manufactured and used in large quantities, due to their useful properties (Breivik et al. 2004a). Small quantities of PCBs can additionally be formed through various combustion processes (Brown et al. 1995). From an environmental perspective, PCBs and PBDEs have created general concern because they are (*i*) toxic (*ii*) persistent (long-lived in the environment) (*iii*) bioaccumulative (thus achieving higher concentrations in living organisms than in the external environment) and (*iv*) semi-volatile (and can thus be transported over long distances in the environment).

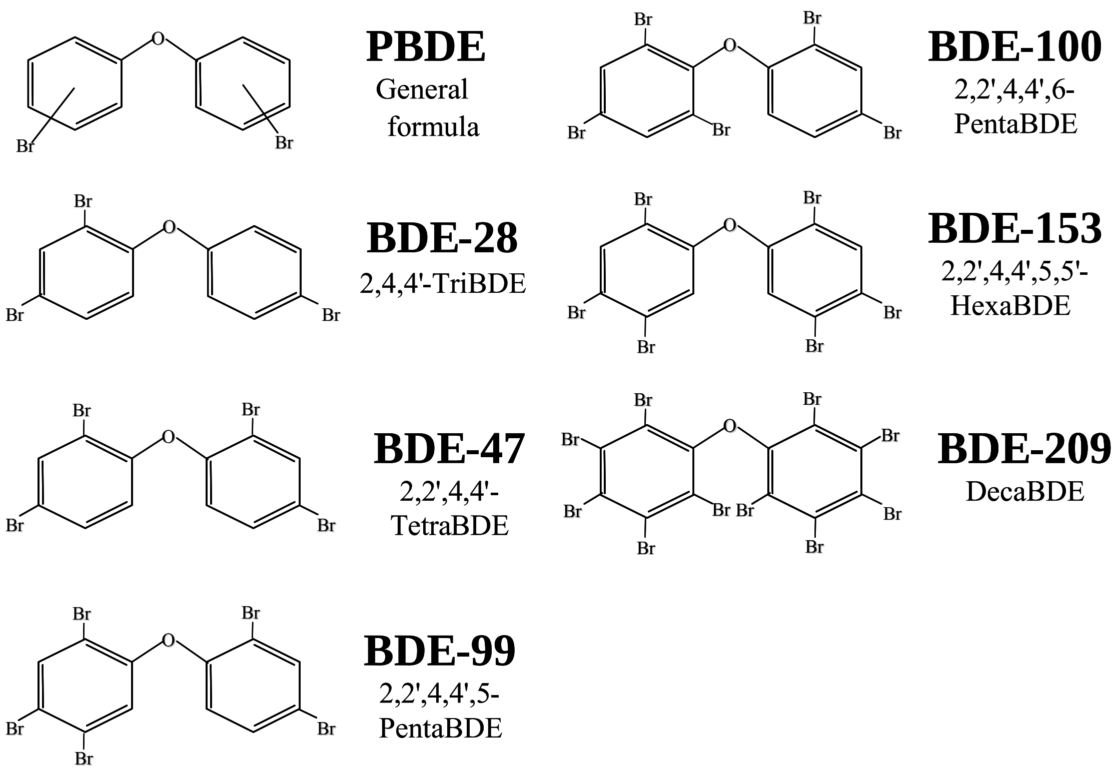

Figure 2. Structural formula for PBDE and six key PBDE compounds (congeners) that are discussed in this report.

Most organic contaminants achieve very low concentrations in air. The atmosphere can still represent an important pathway for contaminants to the aquatic (and terrestrial) environment, where further absorption in the food chain can lead to increased levels that offer ground for concern. For this reason, the project has conducted new measurements of contaminants in air and precipitation to improve understanding of the effect the atmosphere has on levels of selected contaminants in the lake. The main goal of the project was to learn whether atmospheric levels at lake Mjøsa are caused by long-range transport or local atmospheric emissions. Chapter 3 particularly focuses on the importance of the atmospheric burden, and an evaluation of local vs. long-range atmospheric transport of PCBs, PBDEs (BDE-209) and mercury. The project has also conducted new measurements of selected contaminants in rivers and the lake in collaboration with the Norwegian Institute for Water Research (NIVA) and analyzed selected samples from treatment plants in collaboration with Jordforsk – the Norwegian Centre for Soil and Environmental Research (Snilsberg et al. 2005).

A model has also been developed and evaluated during the project (chapter 5) to investigate (*i*) whether the PCB levels in the lake can be explained by the current understanding of existing sources, and (*ii*) whether there are key knowledge gaps related to the sources and behaviour of PCBs in Lake Mjøsa. The report shows that the data on which assessments and calculations are based is limited, and that further investigations should be made to improve knowledge and the basis for decision-making. This is to ensure that any control measure yield the expected environmental benefit. An attempt has therefore been made in the report to identify and discuss critical gaps in knowledge and key areas of uncertainty.

Translation of legend, Figure 3 (following page):

Measuring stations
Passive air samplers
Active air and precipitation samples
Water samples (SPMD)
Sewage plants

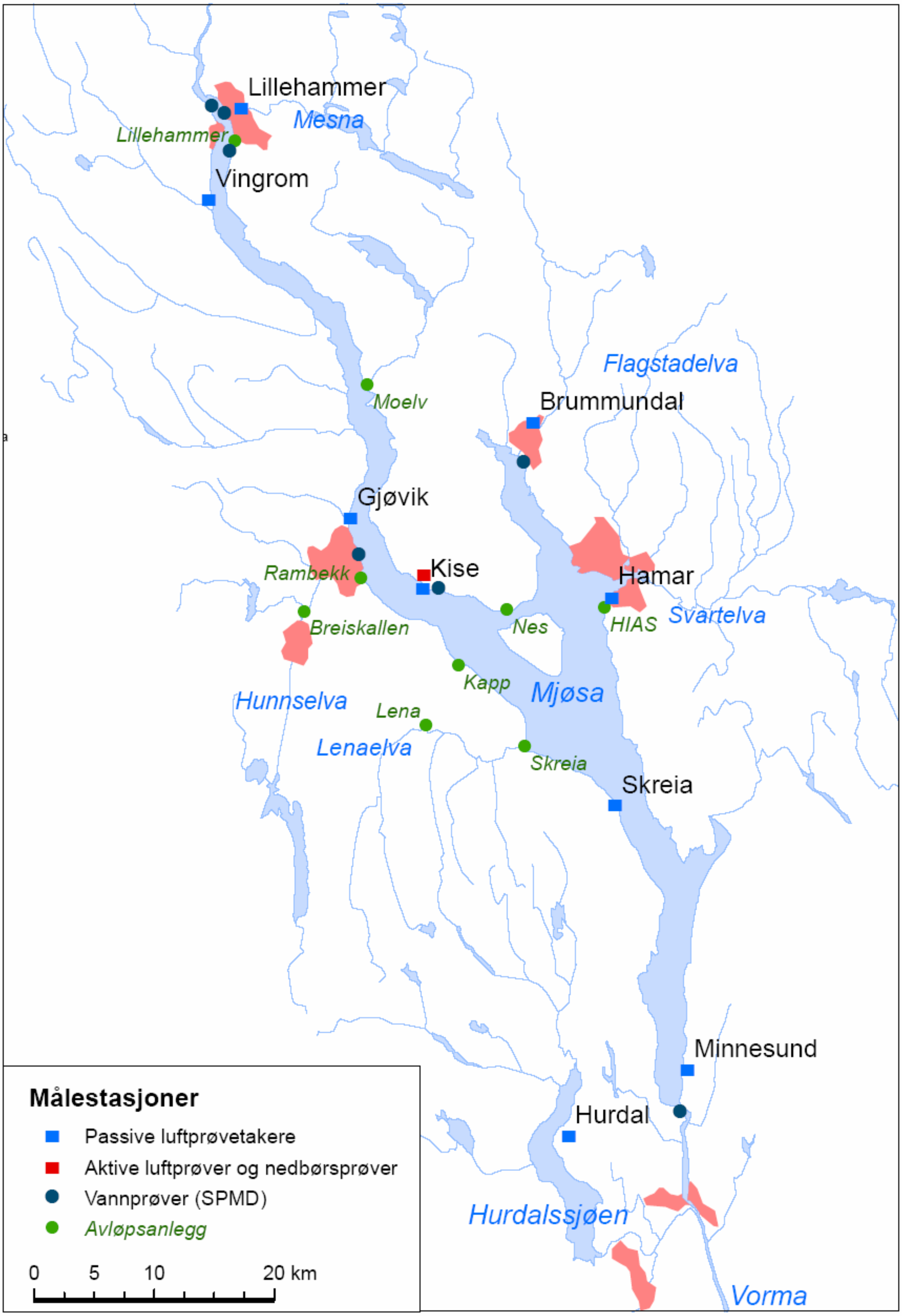

Figure 3. Places in the Lake Mjøsa area where new samples have been collected and analyzed during the project (see the report for a discussion of the results). (Map: Norwegian Pollution Control Authority)

4. Measurements of Selected Contaminants

4.1 Atmospheric levels and sources of contaminants

4.1.1 Air concentrations in Kise

4.1.1.1 Sampling and methods

An active air sampler was placed at the Norwegian Crop Research Institute in Kise in the autumn of 2004 to measure organic contaminants in air. The station was established after consulting meteorological experts regarding the representativity of the Lake Mjøsa region as a whole. These active air samplers consist of a pump where air passes through a filter, followed by a sorbent. The filter will mainly capture organic contaminants that are bound to particles in the air, while the sorbent mainly captures contaminants in the gas phase. 22 samples were collected and later analyzed at the Norwegian Institute for Air Research (NILU) in Kjeller. Each of these samples represent the the air concentration averaged over a two-day period. The date indicated is the start date for the sampling period.

4.1.1.2 PCB results

The results for ΣPCB_7 are shown in Figure 4. ΣPCB_7 denotes the sum of the compounds PCB-28, 52, 101, 118, 138, 153 and 180. The relative distribution of the compounds in each sample is shown in Figure 5. As Figure 4 shows, PCB concentrations in air varied somewhat during the period. There appears to be a general decline after the beginning in early September 2004 up to the end of the year. This is probably due to a drop in temperatures towards winter, as it is known that PCB concentrations in air often vary in accordance with temperature fluctuations (Wania et al. 1998; Haugen et al. 1999), something which can be explained by the increased evaporation of PCBs from surfaces in contact with air at higher temperatures (Wania et al. 1998; Breivik et al. 2004a). By way of comparison, it must be mentioned that the annual average ΣPCB_7 concentration at Lista (based on weekly samples collected over a four-year period) was a factor of 1.4 greater than the seasonal average for samples only collected from September up to and including January (Haugen et al. 1999). In other words, the average concentration of ΣPCB_7 for the samples collected in this study, are likely lower than the annual average value.

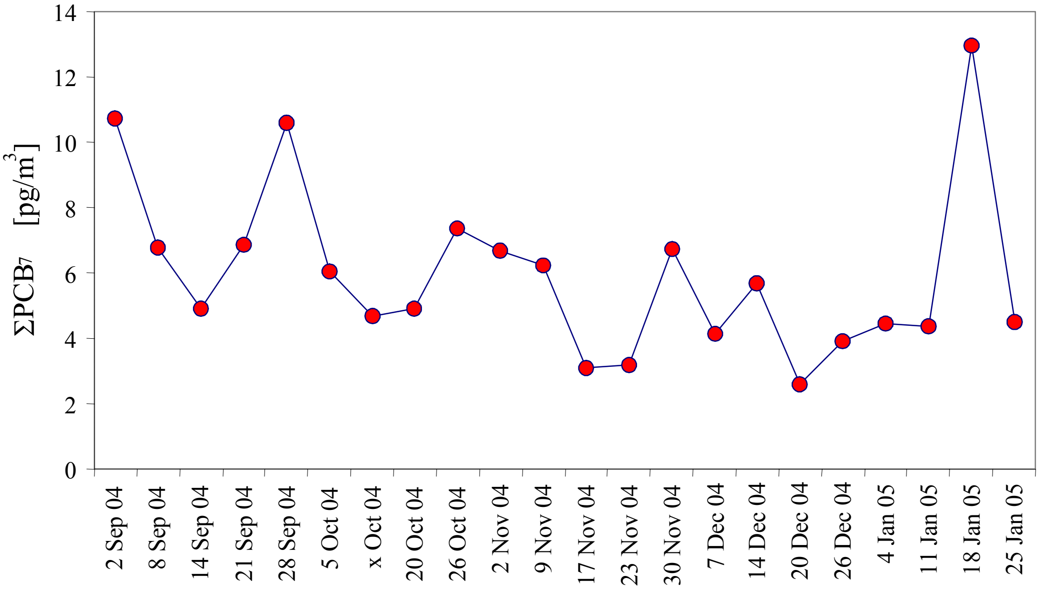

Figure 4. Concentrations of ΣPCB$_7$ in the air at Kise in pg/m^3. Data from September 2004 to January 2005.

The average concentration of ΣPCB$_7$ in air for the period is 6.0 pg/m^3. The highest value was measured on 18 January 2005 (13.0 pg/m^3), while the lowest value was measured on 20 December 2004 (2.6 pg/m^3). Samples that showed higher PCB concentrations will be discussed in greater detail in chapter 4.1.3.

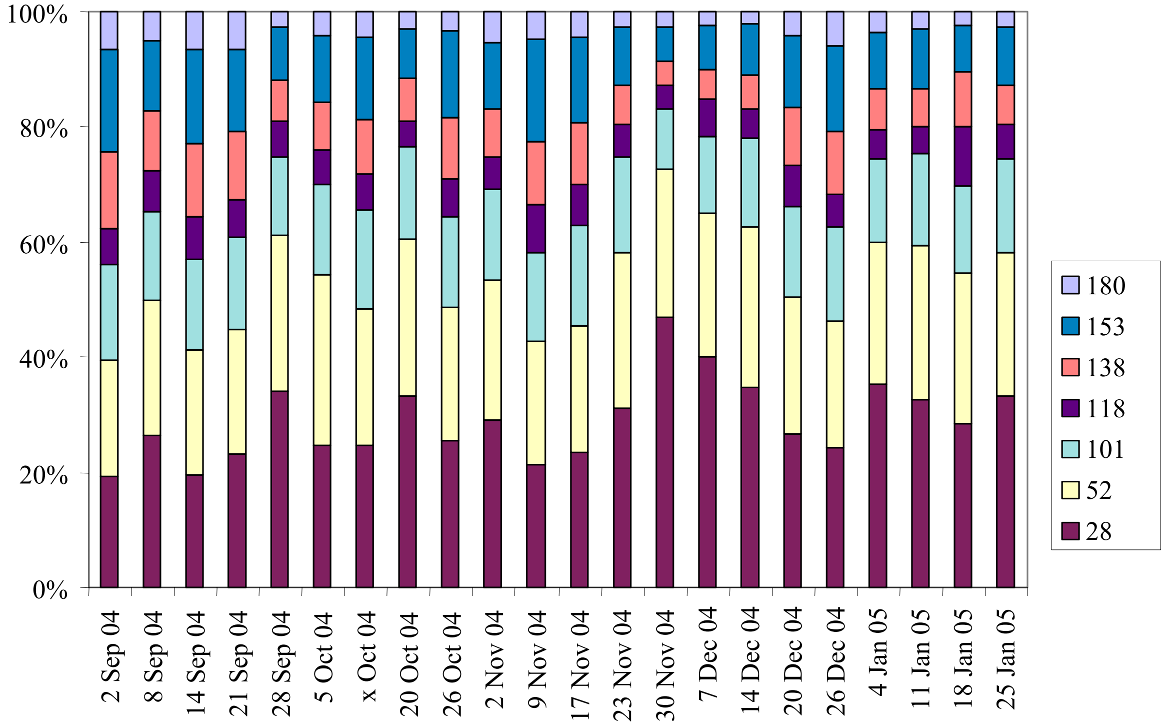

Figure 5. Relative distribution of different PCB compounds in the air samples from Kise (%)[1].

4.1.1.3 ΣPBDE$_5$ results

The results for ΣPBDE$_5$ in the air from Kise are shown in Figure 6. Here ΣPBDE$_5$ is the sum of the compounds BDE-28, 47, 99, 100 and 153. Their relative distribution is shown in Figure 7. ΣPBDE$_5$, more than ΣPCB$_7$, appears to be controlled by episodes with increased levels. While ΣPCB$_7$ varies by a factor of 6, ΣPBDE$_5$ varies by a factor of 50. The average for ΣPBDE$_5$ is 3.5 pg/m^3 during the period, while the maximum (20.5 pg/m^3) and minimum (0.4 pg/m^3) were measured on26 December and 20 October (2004), respectively.

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[1] The exact date for an October sample is missing.

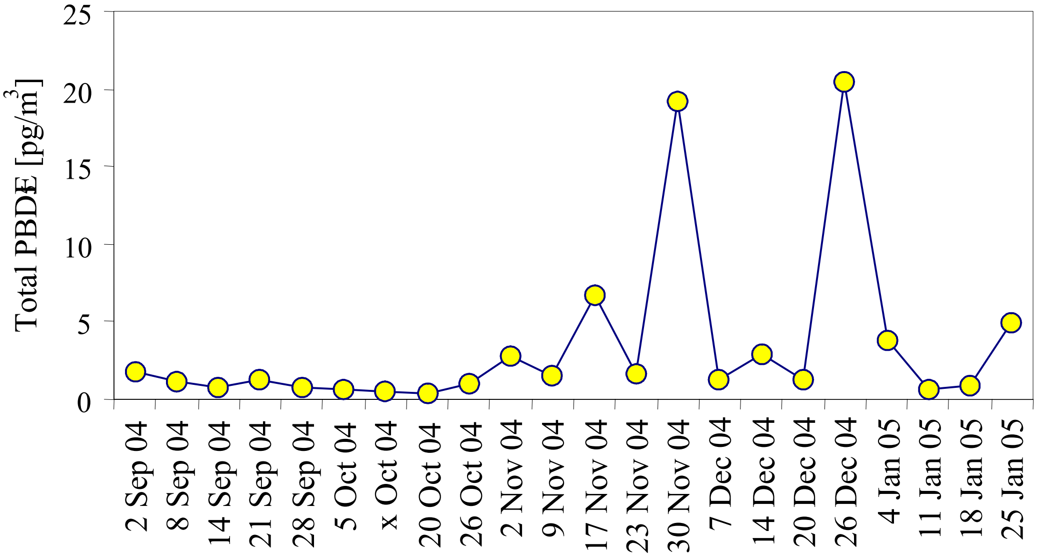

Figure 6. Concentrations of $\Sigma PBDE_5$ in the air in Kise in pg/m^3. Data from September 2004 to January 2005.

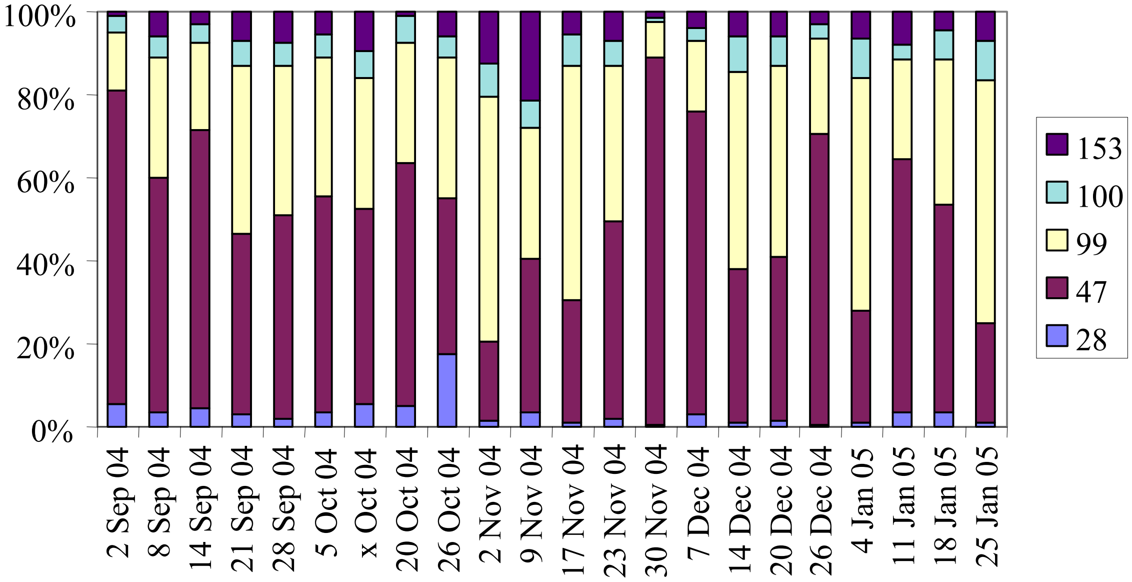

Figure 7. Relative distribution of different BDE compounds in the air samples from Kise (%)[2].

4.1.1.4 BDE-209 results

This project has particularly focused on BDE-209. The results for this compound are shown in Figure 8.

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[2] The exact date for an October sample is missing.

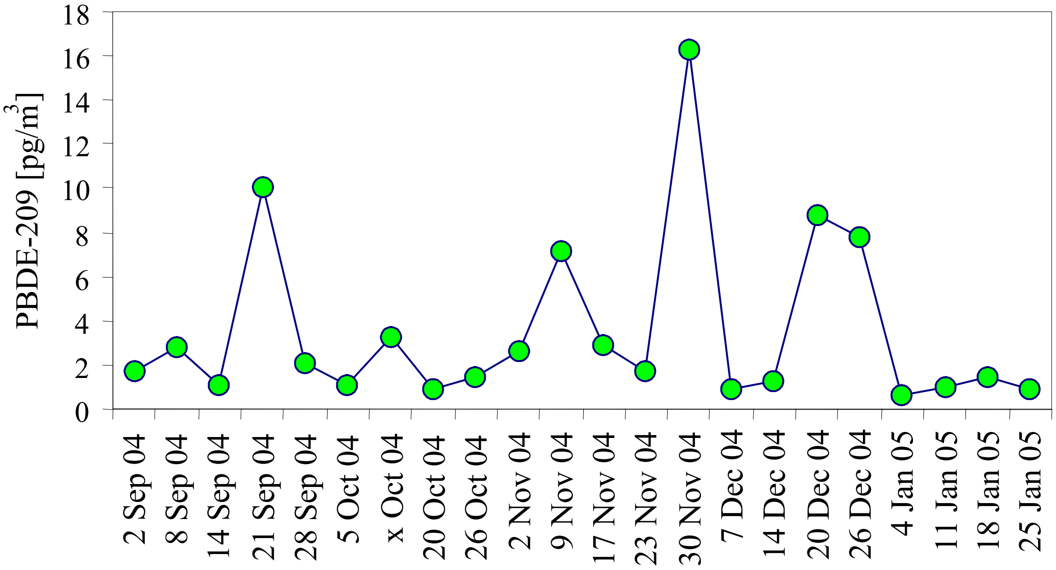

Figure 8. Concentrations of BDE-209 in the air in Kise in pg/m^3. Data from September 2004 to January 2005.

BDE-209 varies by a factor of 24; i.e. more than ΣPCB_7, but less than $\Sigma PBDE_5$. The average for BDE-209 is 3.5 pg/m^3 for the entire period, which is equal to the average for $\Sigma PBDE_5$. The highest concentration of BDE-209 was measured on 30 November 2004 (16.3 pg/m^3), while the lowest concentration was measured on 4 January 2005 (0.7 pg/m^3). The samples that showed elevated concentrations of $\Sigma PBDE_5$ and BDE-209 are discussed in chapter 4.1.3.

4.1.2 Passive air sampling around Lake Mjøsa

4.1.2.1 Sampling and methods

A passive air sampler measures atmospheric levels, integrated over an extended period of time (weeks to years). Several different passive samplers have been described in the literature (Ockenden et al., 2001; Shoeib & Harner, 2002; Wania et al., 2003). They all have in common that they contain sampling material that has a significant affinity and capacity for organic contaminants in air. Secondly, that it is necessary to know the sampler's uptake rate. We have chosen a sampler that uses polyurethane foam (PUF) as sampling material, and which has been tested and evaluated in several studies (see e.g. Shoeib & Harner, 2002; Jaward et al., 2004). This sampler samples an air volume of a couple of cubic meters of air per day (Jaward et al., 2004). For contaminants that are found in both particular form and in a gaseous phase, passive samplers are known to mainly capture the gaseous fraction (Harner et al., 2004). The high-brominated BDE compounds are strongly bound to air particles in the atmosphere (Wania & Dugani, 2003), and therefore are not suitable for studies using passive samplers.

The primary goal of the use of several passive air samplers during this project was to evaluate the spatial variability ofvselected contaminants in the air around Lake Mjøsa. Significant spatial variation indicates variations in emissions of contaminants to air around Lake Mjøsa, while insignificant spatial variation indicates that the burden level is primarily due to atmospheric long-range transport, or that the contaminants in air are well mixed. Passive samplers were deployed from 23 September 2004 until 14–15 December 2004. Ten stations were selected; eight of which were located in the immediate vicinity of Lake Mjøsa (Table 1 and Figure 3). Two stations were chosen to assess how the general burden around Lake Mjøsa can be compared with a location that is presumably subject to a) a heavy burden (Majorstua, Oslo) and b) a presumed background area in the region (Hurdal). For the stations around Lake Mjøsa, we selected potential source areas (towns and villages; stations 5, 7, 8, 10) and stations that would presumably reflect the general background burden (stations 3, 4, 6, 9). Station 6 (Kise) further represents a reference to the active measuring station that was established in Kise (see 1.1.1) in order to evaluate the assumption of its representative localization.

Table 1. Station number, name and coordinates for passive air samplers deployed from 23 September to 14–15 December 2004 (sorted from south to north). Also see Figure 3.

Number	Station name	Longitude	Latitude
1	Majorstua (Oslo)	59˚ 56’ N	10˚ 44’ E
2	Hurdal	60˚ 22’ N	11˚ 04’ E
3	Minnesund	60˚ 26’ N	11˚ 14’ E
4	Skreia A)	60˚ 37’ N	11˚ 07’ E
5	Hamar	60˚ 46’ N	11˚ 05’ E
6	Kise	60˚ 46’ N	10˚ 48’ E
7	Gjøvik B)	60˚ 49’ N	10˚ 41’ E
8	Brummundal	60˚ 54’ N	10˚ 57’ E
9	Vingrom	61˚ 03’ N	10˚ 26’ E
10	Lillehammer B)	61˚ 07’ N	10˚ 28’ E

A) Unfortunately the sample was ruined due to clear-cutting during the sample period. B) These samplers had a decreased opening (whereby the air is allowed to pass) upon collection, compared with when they were deployed. This may have reduced the uptake rate (possible underestimation of atmospheric level).

4.1.2.2 PCB results

The results of the campaign are shown in Figure 9. The results show a significantly higher burden for PCBs in Oslo, compared with the samples from the Lake Mjøsa region. The stations around Lake Mjøsa show a PCB concentration variation of a factor of 3, while the difference between Oslo and the average for the Lake Mjøsa region is a factor of ~50. This is not surprising, as British researchers have shown that the PCB levels in air are 10–100 times higher in urban areas of Europe, compared with the levels in more remote areas. This can be interpreted as clear proof that densely-populated areas are still active sources of PCBs to air (Jaward et al. 2004). Even though this data material is limited, there is little to indicate that the areas selected around Lake Mjøsa should be significant sources of emissions of PCBs to air, compared with Oslo. Still, for Lake Mjøsa as a whole, the results indicate that the burden level is somewhat higher in the cities of Hamar and Gjøvik than in other places. The lowest concentrations of PCBs in air around Lake Mjøsa are observed in Hurdal and Vingrom, while the highest concentrations are detected in the cities of Hamar and Gjøvik. For the last four stations (Lillehammer, Brummundal, Kise and Minnesund), the levels are between 1.5 and 2 ng ΣPCB_7/sample, while the average for the stations around Lake Mjøsa (including Hurdal) is 1.9 ng ΣPCB_7/sample.

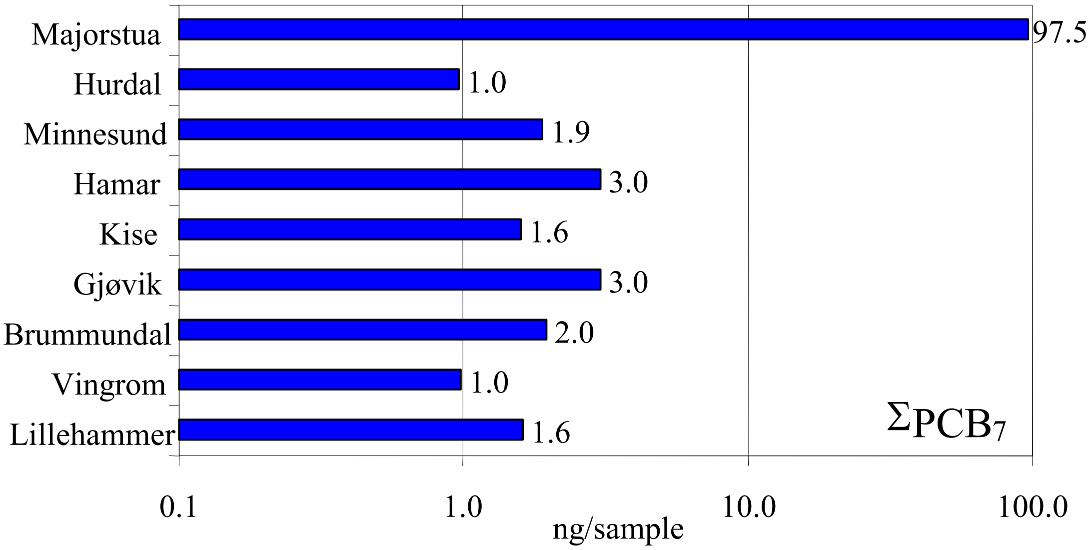

Figure 9. Concentrations of seven selected PCB compounds (ΣPCB_7) on the passive air samplers (ng/sample, logarithmic scale).

4.1.2.3 $\Sigma PBDE_4$ results

For PBDEs, the results in Figure 10 are limited to the total of four compounds (BDE-28, 47, 99, 100), as BDE-153 and BDE-209 are generally strongly bound to air particles for assessment using passive samplers. The highest level of $\Sigma PBDE_4$ was measured in Lillehammer (2.4 ng/sample), while the lowest level was measured in Hurdal (0.09 ng/sample).

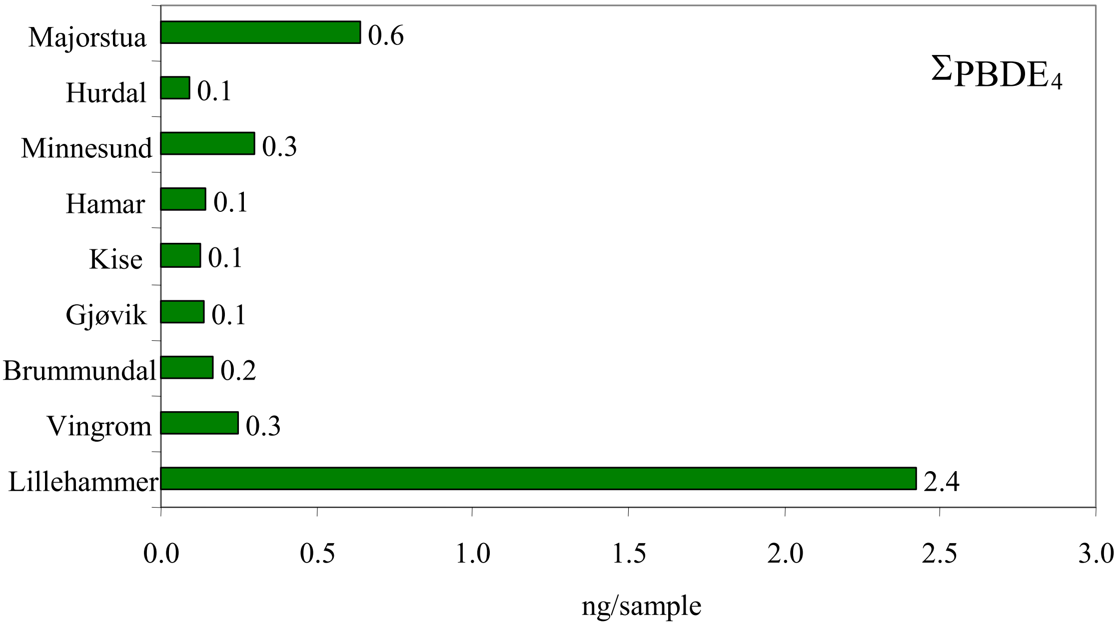

Figure 10. Concentrations of four selected BDE compounds ($\Sigma PBDE_4$) on the passive air samplers (ng/sample).

The difference between maximum and minimum concentrations is thus a factor of 25, which indicates significant spatial variation in the atmospheric burden. Unlike ΣPCB_7, the $\Sigma PBDE_4$ level is higher in Lillehammer than in Oslo. The results also indicate that the $\Sigma PBDE_4$ levels are somewhat higher in Vingrom and Minnesund than is the case for the rest of the stations around Lake Mjøsa (except for Lillehammer). For Vingrom, it is reasonable to assume that this is related to the proximity to Lillehammer. Based on existing data, it is not possible to say which source/s that have led to the increased $\Sigma PBDE_4$ levels in the air in Lillehammer. The burden level in the air in Lillehammer is almost 14 times higher than the average for the other stations around Lake Mjøsa (including Hurdal). The burden level in Lillehammer may be due to a "memory effect" from previous emissions. "Memory effect" means evaporation of PBDEs to air from contaminated media that previously have been exposed to direct emissions/sources (water, sediment, soil). Neither can one exclude that there are one (or more) primary emission sources of PBDEs to air in the Lillehammer region that is/are still to be identified.

4.1.3 Trajectory calculations

Atmospheric trajectory models yield useful information about the origin of air masses. They are calculated using the FLEXTRA model (Stohl et. al., 1995; Stohl & Seibert, 1998) and relies on meteorological data from ECMWF (European Centre for Medium Range Weather Forecasts). The meteorological data that has been used in the model has a spatial resolution of 1.25 degrees and a 6-hour time resolution. This means that for a two-day sampling period (see 4.1.1), 8 trajectory maps can be calculated for each air sample to assess the origin of the air masses. It is important to be aware that FLEXTRA only looks at transport, and neglects processes that influence levels of environmental contaminants in the atmosphere. In other words, processes like degradation of contaminants in the atmosphere and different atmospheric deposition processes that affect the potential for long-range transport have not been taken into consideration. This entails that the relative importance in relation to assessing source contributions has increasing uncertainty as one goes back in time, due to different loss processes (dispersion, dilution, degradation and deposition) that take place during atmospheric transport.

4.1.3.1 PCB results

The three samples with the highest ΣPCB_7 concentrations were taken on 18 January 2005, 2 September 2004 and 28 September 2004, respectively. The lowest concentration was measured on 20 December 2004. For each of these two-day sampling periods we have selected four representative trajectory plots, as shown in Figures 11 A, B, C and D.

18–20 January 2005 (Figure 11A) This is the sample with the highest ΣPCB_7 concentration. Over the two days that the sampler was deployed, the transport pattern was fairly uniform, with air masses transported from Canada over England, via the North Sea and Skagerrak, over Oslo towards Kise. Model studies have proposed that the long-range transport potential for low-chlorinated PCB compounds is generally limited by atmospheric degradation, while high-chlorinated PCB compounds are generally limited by atmospheric deposition processes (see e.g. Wania & Daly, 2002; Wania & Dugani, 2003). This sample had a *relative* ΣPCB_7 composition (see Figure 5) that was high for PCB-118 and low for PCB-153 and PCB-180, which can indicate that the raised ΣPCB_7 level may be due to long-range transport. However, this sample did not have relatively low levels of low-chlorinated PCB compounds. On the other hand, this sample was taken during the winter, when atmospheric degradation is a less effective loss process.

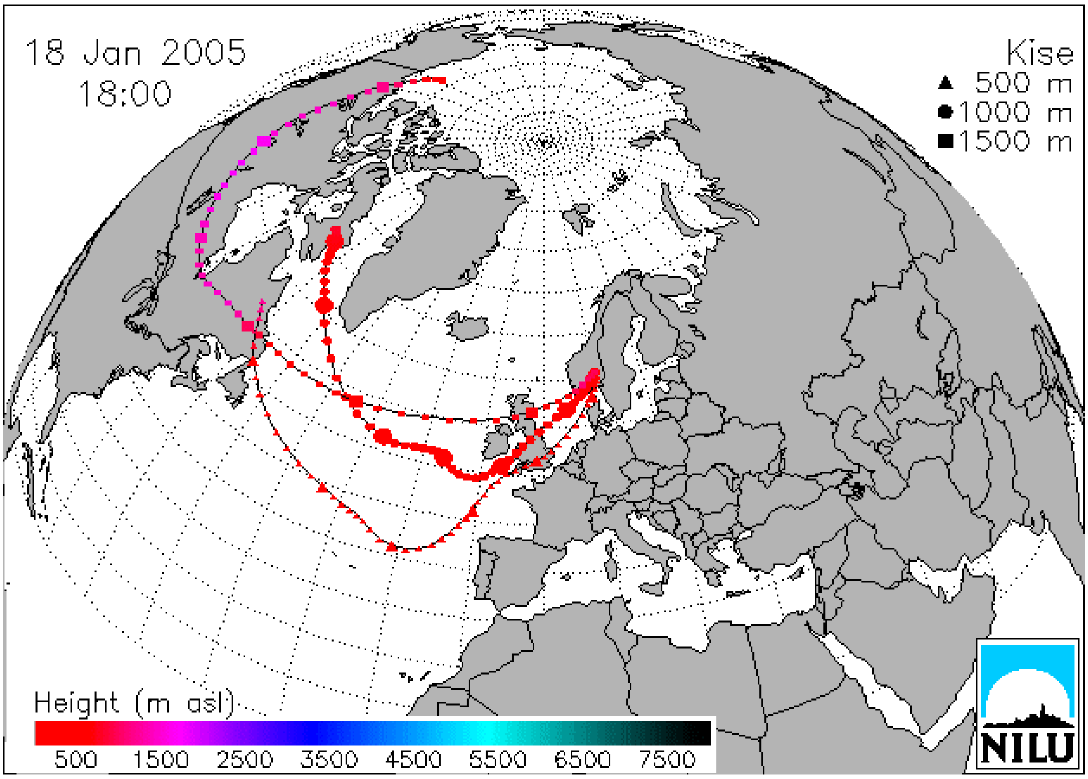

Figure 11A. Trajectory plot for sample showing elevated level of ΣPCB_7.

2–4 September 2004 (Figure 11B) Compared with the previous plot, the air masses have moved at less speed. The general transport pattern during the period indicates that the air masses passed over England, north-western Central Europe and Oslo before arriving in Kise. This sample shows the highest relative levels of high-chlorinated compounds (PCB-138, 153 and 180) and the lowest relative levels of low-chlorinated compounds (PCB-28 and 52) of all samples (see Figure 5), which gives an indication that the increased levels are not due to long-range transport. Seen in the context of the passive air samples, it is natural to assume that the increased PCB levels can be explained by transport from Oslo.

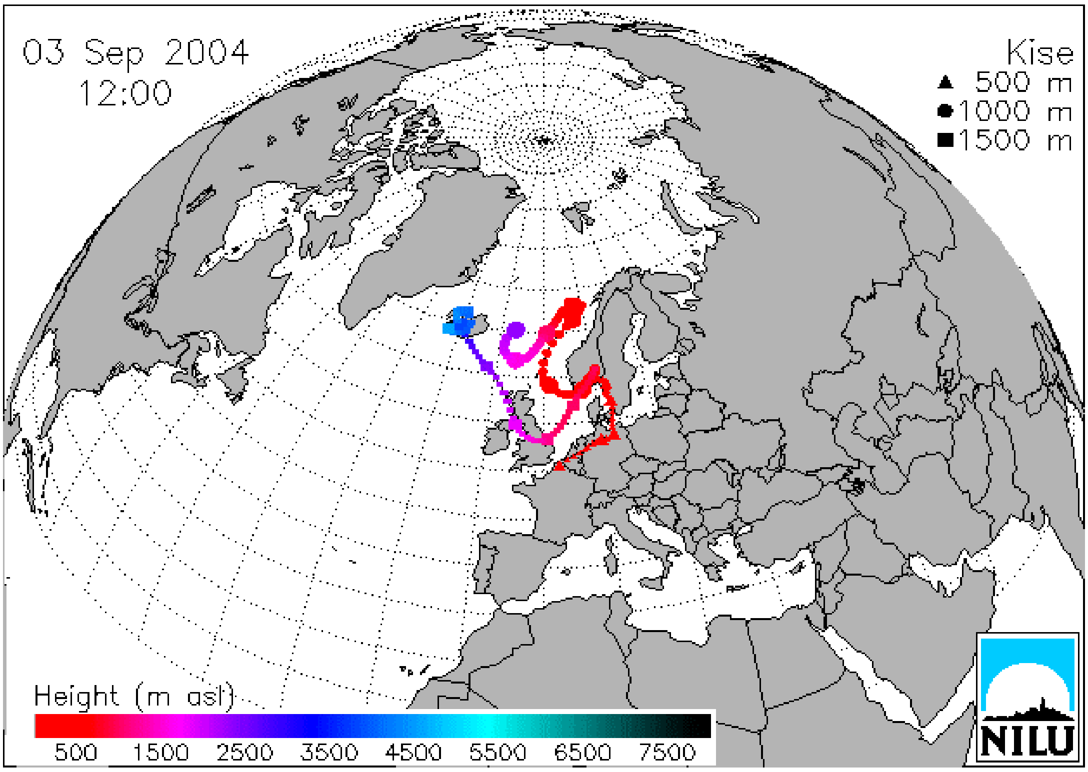

Figure 11B. Trajectory plot for sample showing elevated level of ΣPCB_7.

<u>28–30 September 2004 (Figure 11C)</u> The trajectory map shows a completely different transport pattern, with almost circulating transport around the Baltic Sea. This sample also shows a fairly unique relative composition, with somewhat higher levels of low-chlorinated PCB compounds and somewhat lower levels of high-chlorinated PCB compounds. As suggested earlier, the relatively low level of high-chlorinated PCB compounds may provide an indication of long-range transport (as they are expected to depleted during transport). It can also be mentioned that the PCB compounds that were previously produced in Russia contained smaller quantities of high-chlorinated PCB compounds, compared with the technical PCB mixtures that were produced in other countries (Breivik et al. 2002).

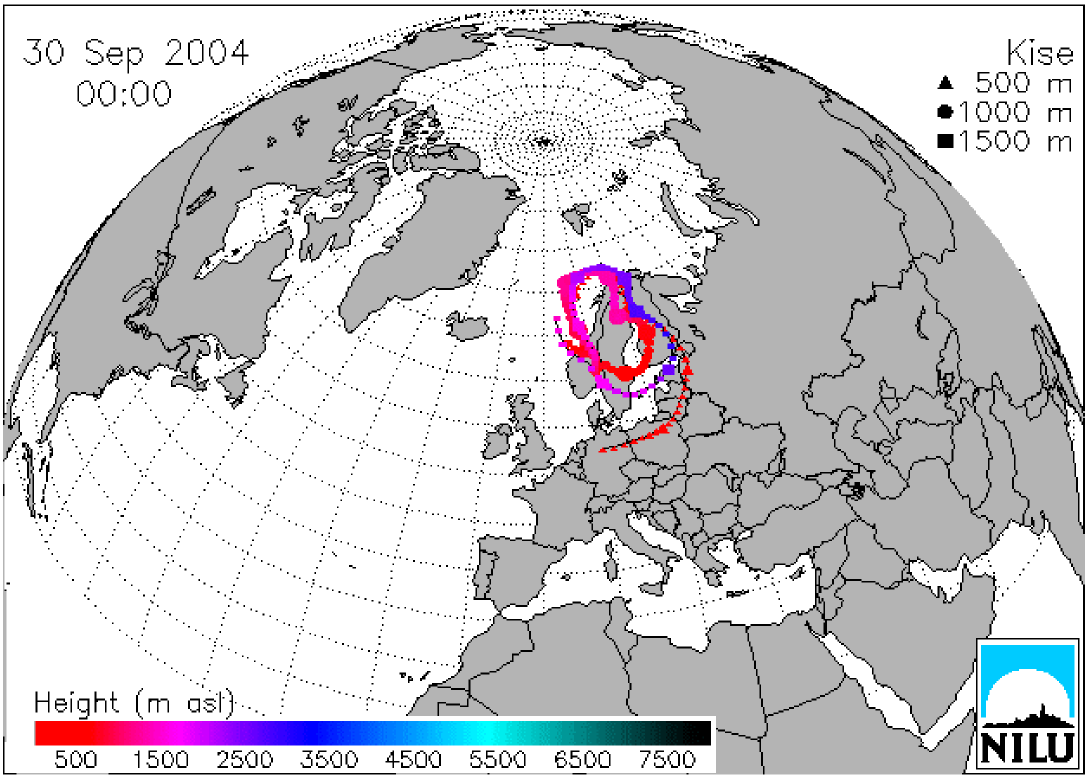

Figure 11C. Trajectory plot for sample showing elevated level of ΣPCB_7*.*

20–22 December 2004 (Figure 11D) This is the sample with the lowest ΣPCB_7 concentration. The air masses generally come from the Arctic regions.

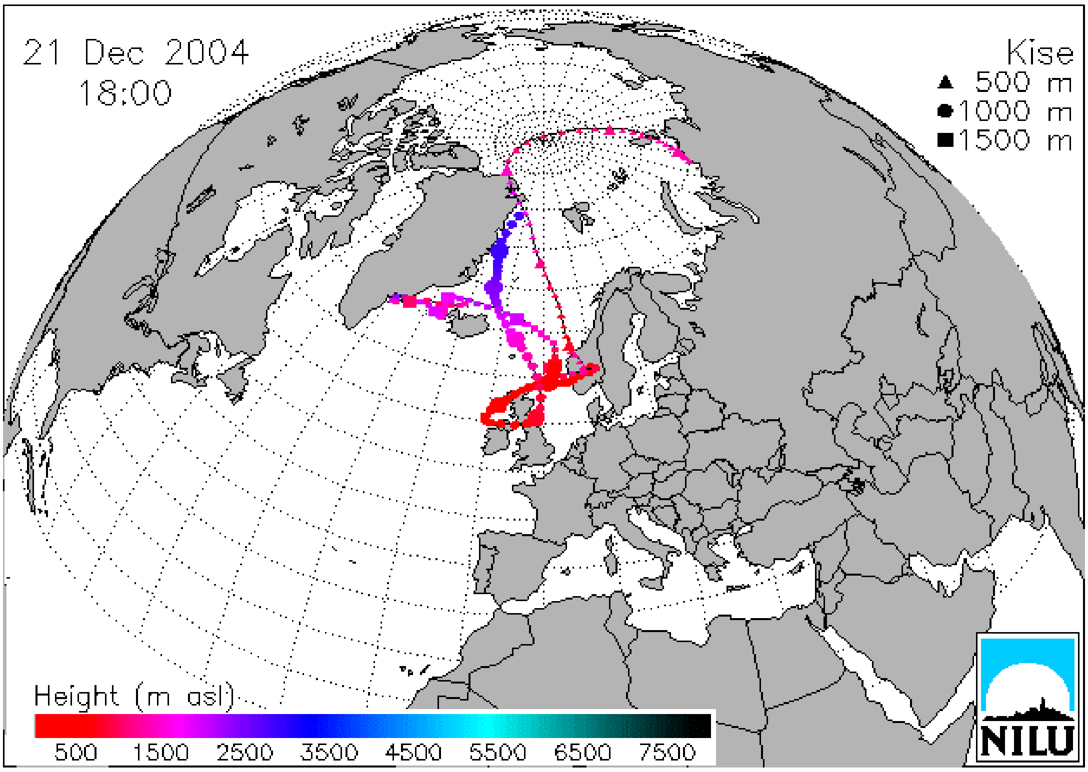

Figure 11D. Trajectory plot for sample showing low level of ΣPCB_7*.4.1.3.2* $\Sigma PBDE_5$ *results*

Wania & Dugani (2003) compared the potential for long-range atmospheric transport of PCBs and PBDEs using four different models. They found out that low-brominated BDE compounds have a long-range transport potential that is comparable to (albeit lower than) that of PCBs, while high-brominated BDE compounds have a more limited potential for atmospheric long-range transport. Hassanin et al. (2004) studied long-range transport of PBDEs by analyzing soil samples (0-5 cm) in Britain and Norway. They learned that the most important components in the technical penta-BDE mixture (BDE-47, -99, -100, -153 and -154) dominated the PBDE composition in soil, and that the PBDE levels were comparable with the PCB levels. They also found that the relative composition of PBDEs in surface soil greatly resembled the relative composition of penta-BDE. This was interpreted as evidence that the transport of compounds that are part of penta-BDEs from sources via air to surface soil show a fairly similar efficacy.

In Kise we find that the clearly highest levels of $\Sigma PBDE_5$ were measured on 26 December and 30 November 2004. The trajectory plot for these two samples can be found in Figures 12A and 12B. It is interesting in itself that neither one of these two samples coincide with the three samples that show raised PCB levels. This provides an indication that the ΣPCB_7 and $\Sigma PBDE_5$ source regions are not the same and/or that the potential for transport varies between these two groups.

26–28 December 2004 (Figure 12A) In the first case, it is natural to assume that the elevated $\Sigma PBDE_5$ levels are mainly influenced by local transport from the Lillehammer region. This is based on the air masses having arrived in Kise from the north, seen in relation to the results from the passive air samplers (Figure 10). Neither can one disregard the possibility that the raised levels to some extent can be due to PBDE sources from central Sweden and Denmark.

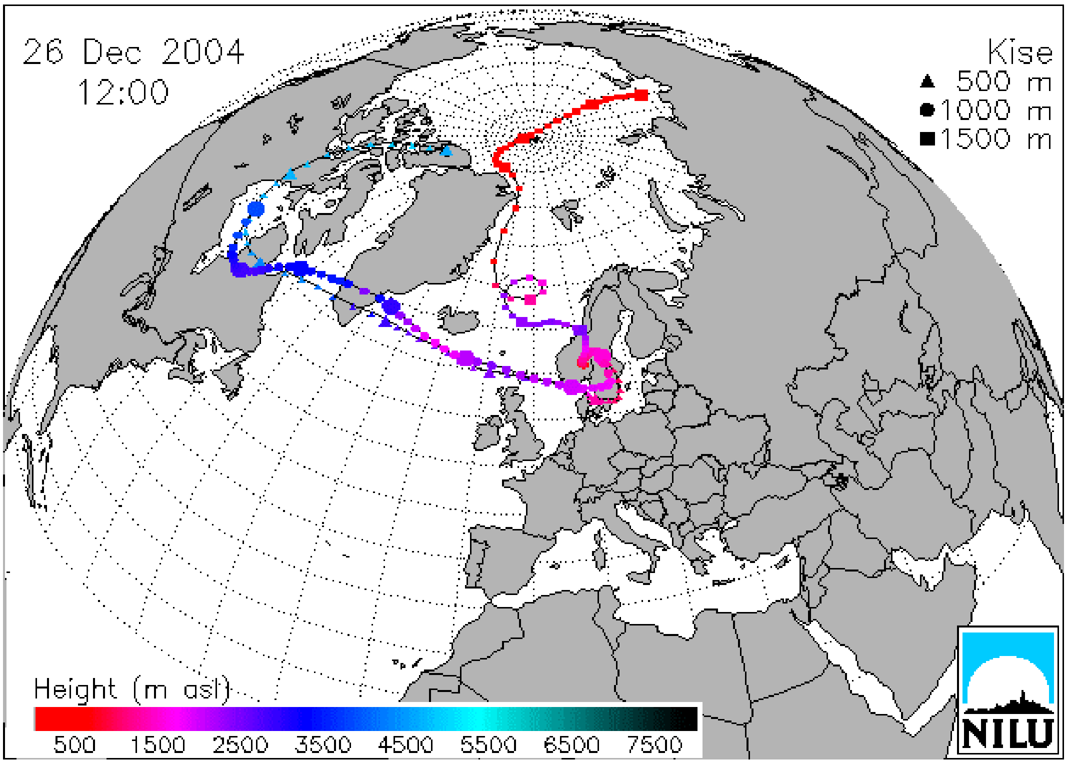

Figure 12A. Trajectory plot for sample showing elevated level of $\Sigma PBDE_5$.

30 November – 2 December 2004 (Figure 12B) The other episode of raised ΣPBDE$_5$ levels shows a different transport pattern. During this two-day period, the situation was fairly stable, with air masses being transported from Britain in over Norway from the southwest, and it thus seems like this episode to a great extent can be explained by long-range transport from Britain. This assumption fits the results from the passive samplers deployed throughout Europe (Jaward et al., 2004). The study by Jaward et al. (2004) found that the PBDE levels in air were significantly higher over Britain than in other parts of Europe (see Figure 7 in Jaward et al., 2004), and that Britain represents a clear regional source of PBDEs to air in Europe.

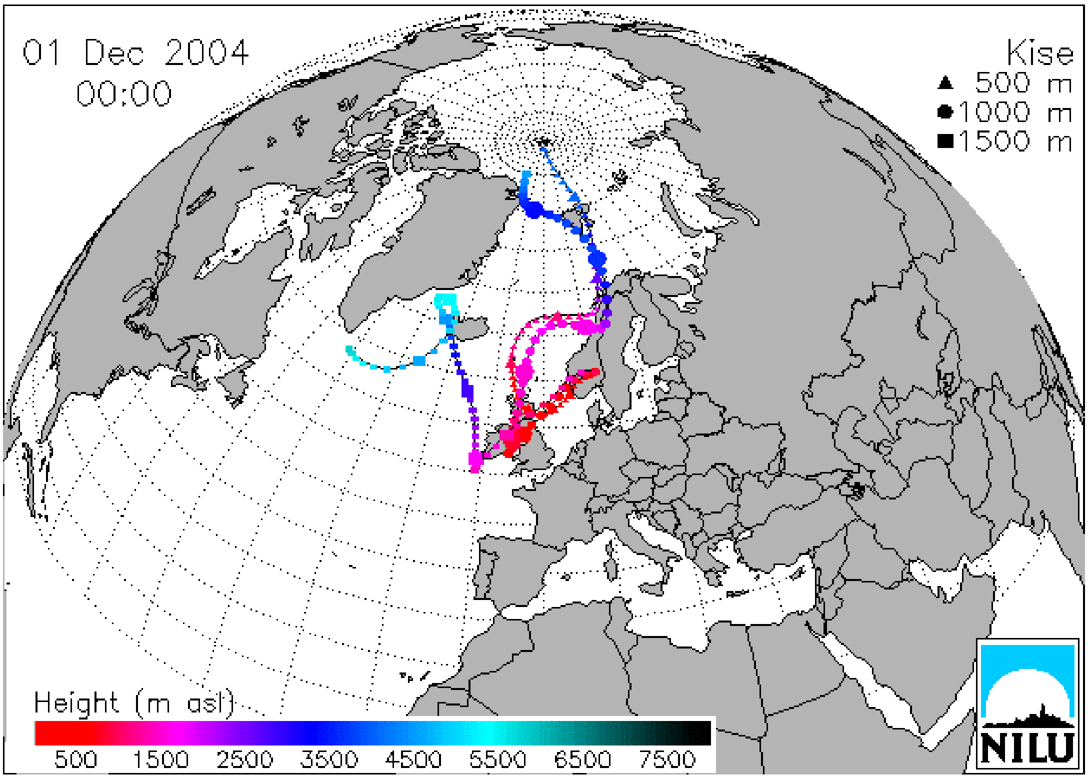

Figure 12B. Trajectory plot for sample showing elevated level of ΣPBDE$_5$

4.1.3.3 BDE-209 results

As mentioned earlier, the average BDE-209 and ΣPBDE$_5$ concentrations in air were almost identical, even though BDE-209 is assumed to have a more limited potential for long-range transport than the components of ΣPBDE$_5$ (Wania & Dugani, 2003). The highest BDE-209 level that was measured in Kise coincides with the likely ΣPBDE$_5$ long-range transport episode from Britain (Figure 12B). BDE-209 is expected to be almost completely bound to particles in air at normal ambient air temperatures. In other words, the long-range potential for BDE-209 is controlled in this case by the long-range potential of air particles, for which this component is bound. Strong winds and dry conditions will favour such long-range transport episodes.

However, episodes characterized by local atmospheric transport cannot be disregarded. The sample with the third-highest BDE-209 concentration is the same one as the one with elevated ΣPBDE$_5$ levels (Figure 12A), while the trajectory plot for the sample with the second-highest BDE-209 concentration from 21–23 September 2004 can be found in Figure 13. On the other hand, this episode showed somewhat more variable transport patterns during the sampling period, where the air masses occasionally arrived from the northwest, and at times directly from the southwest after passing over Britain.

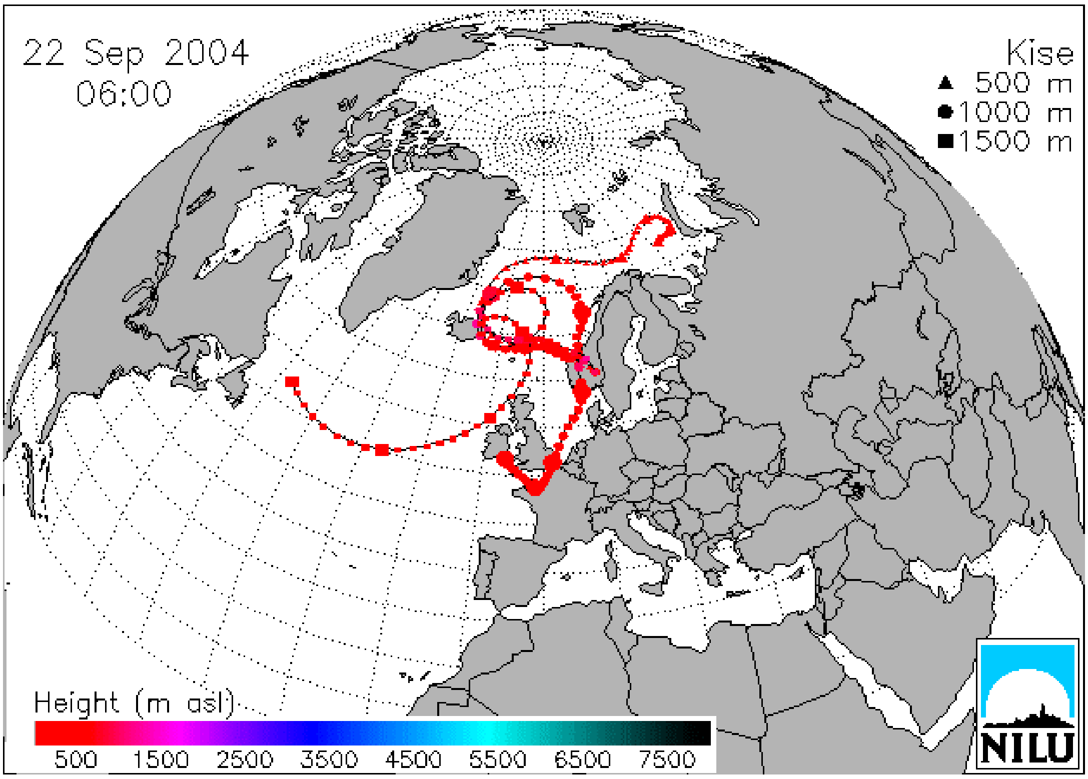

Figure 13 Trajectory plot for sample showing elevated level of BDE-209.

4.1.4 Hg sources through precipitation

4.1.4.1 Sampling and analysis

Samples of mercury in precipitation were collected in Kise. IVL's (IVL Swedish Environmental Research Institute) mercury bulk sampler was used for precipitation sampling. The precipitation sampler is made of glass, and was placed 2 metres above the ground. The samples were (mainly) collected on a monthly basis, and later analyzed at NILU's laboratory in Kjeller. In the figures, the date indicates the start date for the sampling. One sample ends when the next one begins. In December, February and April there was not enough precipitation for an analysis to be conducted.

The mercury in precipitation was analyzed using a fully-automatic "Tekran 2600" instrument. This method reduces all of the mercury in the sample to Hg° using stannous chloride, and it is then sorbed on a gold trap. Further in the analysis, the Hg° was heat-desorbed and detected using atomic fluorescence spectrophotometry. The detection limit for the method is 0.2 ng mercury in absolute quantity.

4.1.4.2 Results

A total of six samples were taken from 17 September 2004 up to and including 3 June 2005. The highest concentration (28 ng/l) was measured from 17 September to 1 October, while the lowest concentration (5.7 ng/l) was measured in January (Figure 14). The sample medians are 9.9 ng/l. The mercury deposition in Kise for this six-month period is 3.8 µg/m^2 (Figure 15). As these are monthly samples, the results cannot be related to the air masses' origin during the sampling period. The results have been compared and evaluated against results from other studies in chapter 4.1.5.2.

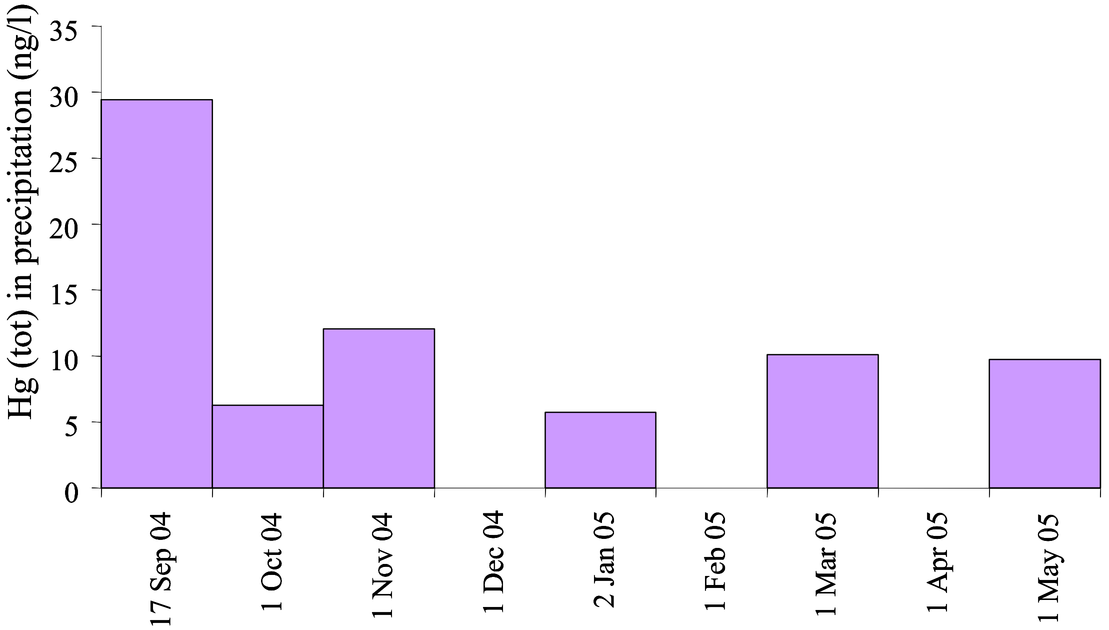

Figure 14. Concentration of total mercury in precipitation, Kise.

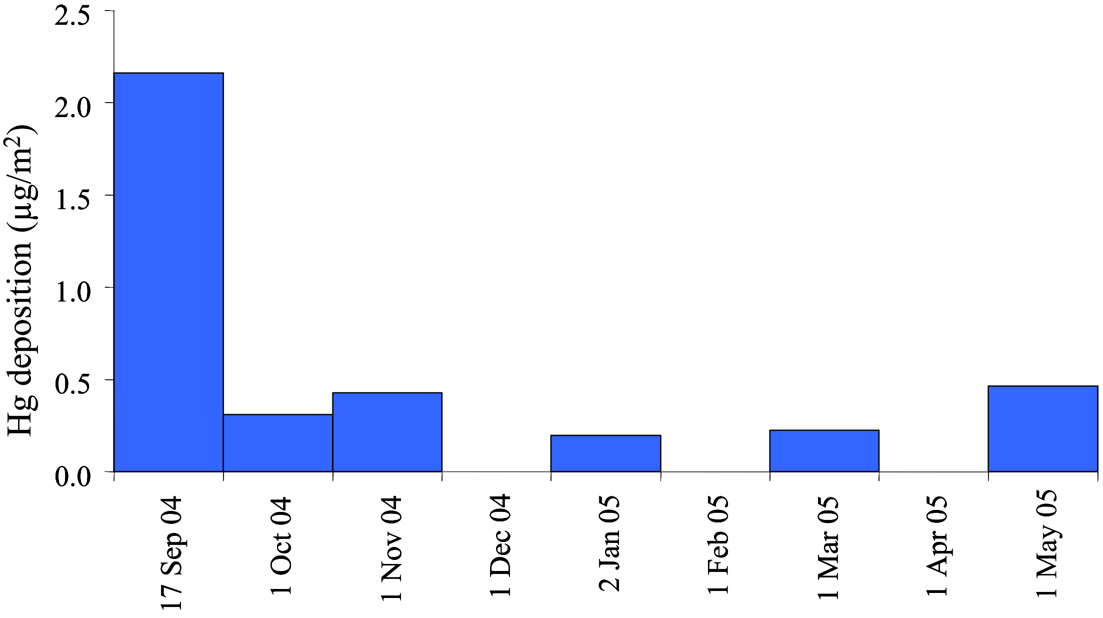

Figure 15. Deposition of total mercury in precipitation, Kise.

4.1.5 Concluding assessment of local vs. long-range atmospheric transport

4.1.5.1 PCBs and PBDEs

Table 2 summarizes key results from the project's air measurements. It must be pointed out that the data basis is limited, and that only trends in the data material can be evaluated. A continued monitoring of organic contaminants in air in the future will be able to strengthen the statistical power in relation to assessment of further control measures. The Kise (active) air measurements still show clearly that the variability in concentrations *over time* is far less significant for PCBs than for PBDEs. This shows that PCBs are far more mixed in the atmosphere around Lake Mjøsa than is the case for PBDEs. This gives a further indication that it will probably be a greater challenge to both identify and implement efficient control measures (either locally, nationally or internationally) in order to reduce the general PCB burden in the air around Lake Mjøsa, compared with PBDEs. The trajectory calculations and the passive air samplers still show that elevated PCB levels appear to both be due to 'local' transport from Oslo, and long-range transport from other countries. The passive samplers also provide an indication of a somewhat higher PCB burden level in connection with the cities of Hamar and Gjøvik than is the case for the other stations around Lake Mjøsa.

Table 3 compares PCB data from Kise with data from other places in Norway. The PCB burden in Kise is almost identical with the one measured in Birkenes in southern Norway, which is presumed to be a typical background level, while Oslo shows a level that is about 10 times greater.

For PBDEs it appears that further measures to identify and reduce emissions to air near Lillehammer can have a beneficial effect on the local atmospheric burden. The effect of the increased atmospheric burden near Lillehammer on PBDE levels in the lake should also be investigated further. There are also strong indications that episodes of long-range transport can lead to periods with a higher burden, where Britain appears to be a probable source region for PBDEs on a European scale. It is also worth pointing out that the BDE-209 concentration level is directly comparable with ΣPBDE$_5$, based on the measurements from Kise.

Table 2. Some key results from the air measurements [A].

	Active (Kise, n=22) pg/m^3			Passive (n=9) ng/sample [B]		
	average	range	max/min	average	range	max/min
PCB-28	1.7	0.7–3.7	5	4.2	0.2–33.6	142
PCB-52	1.5	0.6–3.4	6	4.7	0.2–38.3	171
PCB-101	0.9	0.4-2.0	5	2.0	0.2-15.3	99
PCB-118	0.4	0.2–1.4	8	0.5	0.1–3.5	50
PCB-138	0.5	0.2–1.4	7	0.4	0.1–2.4	32
PCB-153	0.7	0.3–1.9	6	0.6	0.1–3.7	29
PCB-180	0.2	0.1–0.7	8	0.1	0.03–0.7	21
ΣPCB$_7$	6.0	2.6–13.0	6	12.5	0.97–97.5	101
BDE-28	0.04	0.01–0.16	11	0.04	<0.004–0.15	35
BBDE-47	2.1	0.22–17.0	79	0.28	0.06–1.38	24
BDE-99	1.0	0.12–4.7	40	0.13	0.03–0.70	22
BDE-100	0.2	0.02–0.76	31	0.03	<0.001–0.19	276
ΣPBDE$_4$	3.3	0.4–19.9	50	0.48	0.09–2.42	26
BDE-153	0.2	0.01–0.61	122	NA	NA	NA
ΣPBDE$_5$	3.5	0.4–20.5	51	NA	NA	NA
BDE-209	3.5	0.7–16.3	24	NA	NA	NA

[A] In the passive air sample from Hurdal, BDE-28 and BDE-100 were below thelimit of detection (LOD). In the calculations, they have been assigned a value of ½ LOD. [B] Passive samplers are not suitable for determining high-brominated BDE compounds (BDE-153 and BDE-209).

Table 3. Comparison of PCB concentrations in air from this study (Kise) with other locations in Norway (Oslo, Birkenes, Ny-Ålesund).

	Kise [A]	Oslo [B]	Birkenes [C]	Ny-Ålesund [C]
PCB-28	1.7	11.9	1.6	2.0
PCB-52	1.5	16.2	1.3	1.0
PCB-101	0.9	10.8	0.8	0.4
PCB-118	0.4	3.6	0.3	0.1
PCB-138	0.5	4.0	0.4	0.1
PCB-153	0.7	6.1	0.6	0.2
PCB-180	0.2	1.6	0.2	<0.1
ΣPCB$_7$	6.0	54.2	5.3	3.9

[A] This study, n=22. [B] Oslo, 2002, n=9 (Breivik et al. 2004b). [C] Birkenes, 2004, n=52 and Ny-Ålesund, 2004, n=52 (Aas et al., 2005)

4.1.5.2 Mercury

When the mercury data from Kise is compared with corresponding data from Birkenes, Aust-Agder county, it can be seen that the mercury concentrations in precipitation are somewhat higher in Kise than in Birkenes. When comparing months where both stations have yielded sufficient precipitation for analysis, the medians are respectively 9.9 and 6.5 ng/l (Fig. 16). However, as the precipitation volume is significantly lower in Kise than in Birkenes, the mercury deposition is almost twice as high in Birkenes as in Kise (respectively 7.1 and 3.8 µg/m^2) (Fig. 17) for this six-month period. Over a one-year period, the normal precipitation volume in Kise is 585 mm, while it is 1200 mm in Birkenes. It must be pointed out that the data material is limited (only six samples), and this will therefore only be an indication in the data material. If one presumes that the precipitation samples for mercury in Kise are representative of the entire year, the annual deposition will be approximately 5.3 µg/m^2 year. If we further assume that Kise is representative of the entire Lake Mjøsa region, mercury to Lake Mjøsa through precipitation will be approximately 2 kg annually. The total annual deposition for six CAMP[3] stations was in the range 6–10 mg/m^2 per year in 1999–2002. The annual precipitation at these stations was 600–1200 mm.

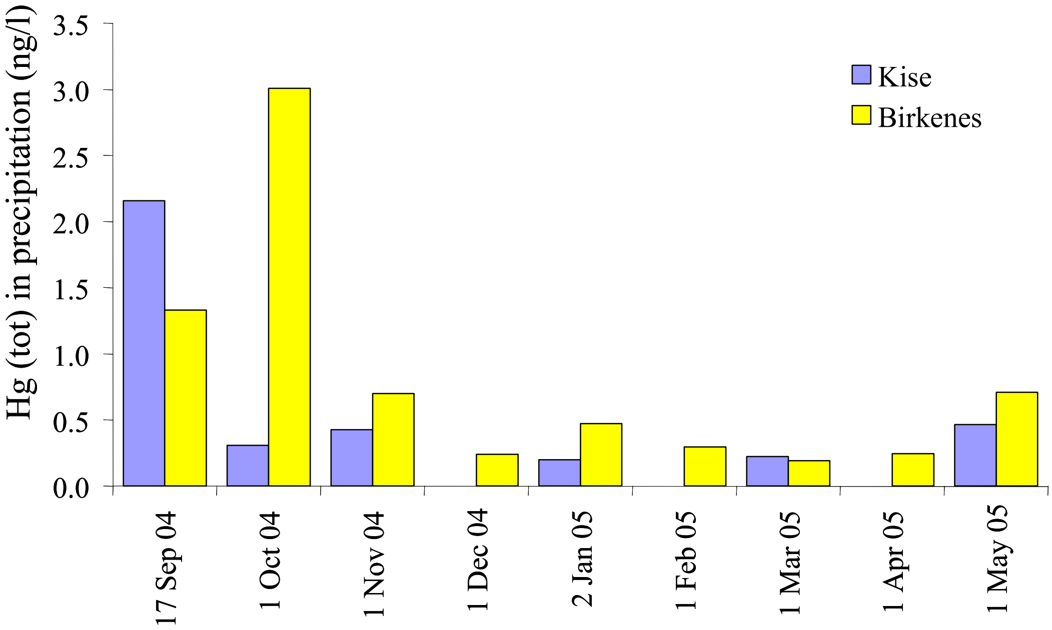

Figure 16. Concentration of total mercury in precipitation, Kise and Birkenes.

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[3] Comprehensive Air Monitoring Programme

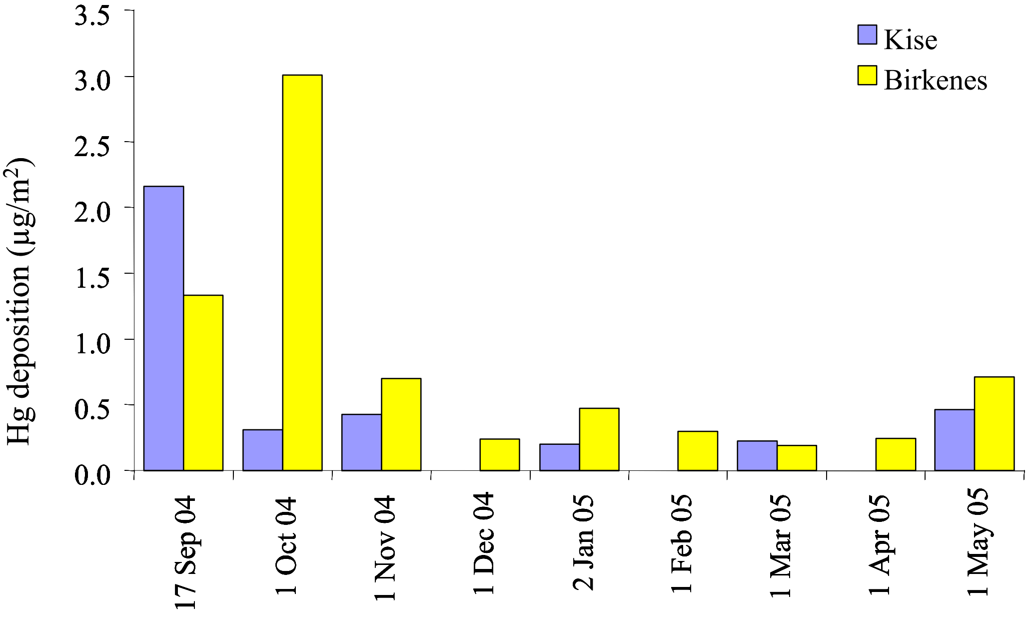

Figure 17. Deposition of total mercury in precipitation, Kise and Birkenes.

Data from EMEP[4] shows that Norwegian anthropogenic emissions of mercury to air are approximately 37% of the amount Norway recieves from other countries (Berg et al. 2003). Data from Statistics Norway shows that there have not been any significant emissions to air of Hg from local sources in the Lake Mjøsa district (Berg et al. 2003). This means that there are no industrial enterprises in control classes 1, 2 and 3, which all have an annual duty to report their emissions to air to the authorities. However, small industries in control class 4 have no duty to report (Berg et al. 2003). Therefore there may be active sources around Lake Mjøsa that can affect local atmospheric sources to Lake Mjøsa.

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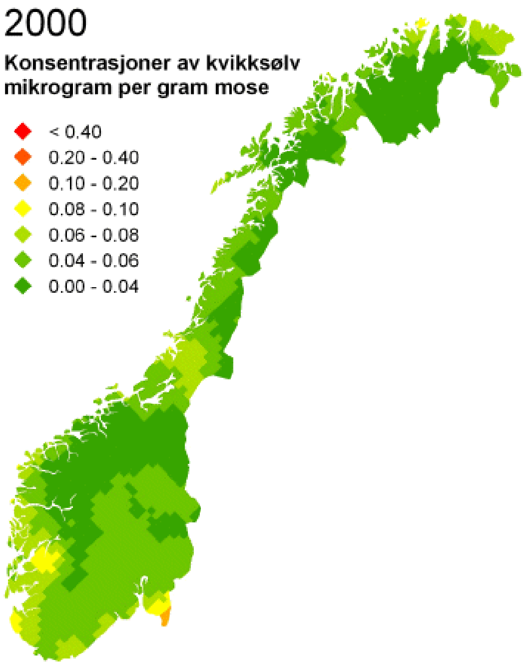
[4] Steering Body to the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmissions of Air Pollutants in Europe.



Translation of legend: Concentrations of mercury, microgram per gram moss

Figure 18. Concentration of mercury in moss.

Neither do the results of the moss studies in 1985, 1990, 1985 and 2000 (Berg et al. 2003) show that the Lake Mjøsa area is a mercury hotspot in relation to airborne sources (see Figure 18). The average concentration for samples taken in the Lake Mjøsa area (Table 4) shows an average concentration of some 0.38 µg/g, while the average concentration for the entire country (500 samples) is 0.55 µg/g. All of the moss samples were collected at least 300 m from main roads and populated areas, and at least 100 m from small roads and detached houses. The possibility of the concentrations being higher in certain areas cannot be disregarded.

Table 4. Moss samples from the national study taken near Lake Mjøsa (Berg et al. 2003).

Location	Longitude	Latitude	Hg (µg/g)
Mesnali	61˚03’ N	10˚50’ E	0.39
Vingrom	60˚03’ N	10˚17’ E	0.49
Synnfjell	61˚07’ N	9˚43’ E	0.35
Øyer	61˚18’ N	10˚25’ E	0.46
Svatsum	61˚20’ N	9˚45’ E	0.30
Ringebu	61˚33’ N	10˚10’ E	0.30

4.2 Levels in Lake Mjøsa and riverine inputs

4.2.1 Levels in Lake Mjøsa

4.2.1.1 PCB results

The determination of organic contaminants in water represents a significant analytical challenge, due to very low concentrations in the water phase. As is the case for active air samples, a significant volume is required for exact determination. A total of four samples, each exceeding 250 l, were collected by NIVA (the Norwegian Institute for Water Research) on 9 and 10 September 2004. Table 5 shows the PCB results, corrected for field blank values. The samples were taken outside Kise at depths of 5 and 25 m.

Table 5. PCB and PBDE concentrations in water outside Kise on 9 and 10 September 2004 (in pg/L).

	9/9-2004 5 m	9/9-2004 25 m	10/9-2004 5 m	10/9-2004 25 m
PCB-28	0.87	2.50	2.31	2.93
PCB-52	0.96	3.06	2.75	3.18
PCB-101	0.29	0.79	0.73	0.99
PCB-118	0.10	0.28	0.34	0.54
PCB-138	0.09	0.22	0.38	0.64
PCB-153	<0.41	0.08	0.29	0.78
PCB-180	0.06	0.08	0.07	0.16
ΣPCB_7	2.36 [A]	7.02	6.86	9.22
BDE-209	< 22.9	< 21.9	< 21.7	< 18.0

[A] The PCB-153 concentration has been set at equal to 0 in the ΣPCB_7 calculation.

The results from the four samples show that the levels were generally lower in the upper layers of the lake during the period in which the samples were taken. They also show that the concentration was lower on 9 September, compared with 10 September. For BDE-209, all of the blank-corrected samples were below the detection level.

Table 6. PCB concentrations (pg/sample) in SPMDs at different stations (exposed from 7/9 September to 21/22 October). The results from Lillehammer, Kise and Minnesund have been corrected against blank values for the respective stations, while the other stations have been corrected in terms of the average of the blank values from these three stations. For Minnesund, Lågen, Mesna and Hunnselva the results have been presented as the average of the two parallel samples.

PCB	Lille–hammer	Kise	Furnes–fjorden	Minne–sund	Lågen	Mesna	Hunns–elva
PCB-28	368	467	561	256	191	175	2198
PCB-52	253	541	706	346	157	268	1755
PCB-101	197	452	643	287	145	353	1331
PCB-118	103	203	355	137	68	210	683
PCB-138	114	226	306	161	92	207	717
PCB-153	163	291	379	614	118	245	620
PCB-180	35	42	98	35	22	54	155
ΣPCB_7	1233	2221	3049	1837	793	1511	7459

The four water samples that were taken outside Kise were supplemented with passive SPMD samplers that were deployed at four stations in the Lake Mjøsa area (Lillehammer, Kise, Furnesfjorden and Minnesund) and in three rivers (Lågen, Mesna, Hunnselva). As was the case with the passive air samplers, SPMDs yields a semi-quantitative picture of the concentration level of organic contaminants in water. They are therefore a particularly useful tool for evaluating spatial variations. The method cannot be applied to the determination of organic contaminants that are bound to particles; BDE-209 has consequently not been included in this analysis. Recordings over time were studied for selected stations, but in comparing results between stations, only results from stations with almost identical exposure time are shown in Table 6.

The results indicate that the PCB level in Lillehammer is the lowest of the four stations in the Lake Mjøsa area. This can be explained by the consistently lower PCB levels in the Lågen. The levels in Kise and Furnesfjorden are higher than in Lillehammer, and indicate possible local sources and transport of PCBs in the vicinity of these areas. As mentioned earlier, the PCB levels in air are somewhat higher around the cities of Hamar, Gjøvik and Brummundal than in the other stations around Lake Mjøsa (see Figure 9). Hunnselva also has the highest PCB concentrations, based on the SPMD analyses, which may possibly help explain the elevated PCB levels in Kise. Further, elevated PCB concentrations in sediments near Hamar have been reported earlier (Kjellberg and Løvik, 2000).

4.2.2 Riverine inputs to the Lake

Jordforsk (Snilsberg et al. 2005) recently published a report assessing sources of different contaminants to the Lake Mjøsa region from landfills and treatment plants. Jordforsk (Snilsberg et al. 2005) also analysed levels of selected contaminants in several rivers (Svartelva, Flagstadelva, Mesna, Hunnselva and Lenaelva). However, the sample volume was too limited for a specific determination of PCBs and BDE-209. All of the PCB samples and two of the BDE-209 samples were below the detection level (see Table 7). They therefore have not been discussed.

Table 7. BDE-209, PCB and Hg concentrations from five rivers (in ng/L).

Concentrations in rivers (ng/L)										
Treatment plant	BDE-209	PCB-28	PCB-52	PCB-101	PCB-118	PCB-138	PCB-153	PCB-180	ΣPCB_7	Hg
Svartelva	0.27	<0.02	<0.01	<0.02	<0.01	<0.03	<0.03	<0.02	<0.14	<5
Flagstadelva	0.23	<0.03	<0.02	<0.02	<0.01	<0.02	<0.02	<0.01	<0.13	<5
Mesna	0.65	<0.01	<0.03	<0.07	<0.05	<0.04	<0.04	<0.01	<0.24	<5
Hunnselva	<0.08	<0.02	<0.02	<0.02	<0.01	<0.03	<0.03	<0.02	<0.13	<5
Lenaelva	<0.15	<0.02	<0.02	<0.02	<0.01	<0.01	<0.02	<0.01	<0.11	<5

4.2.3 Mercury

There have been several mercury sources to the Lake Mjøsa region (Bekkevold *et al.* 1988). The most important ones were emissions from the wood-processing industry (Mesna cardboard factory in Lillehammer). During the ten years up to 1970, about 2.5 tonnes of mercury were released. In addition, mercury was fed through the drainage to Lake Mjøsa and the rivers from fields where stained seed was used (Nashoug *et al.* 1999). These emissions have now been stopped. In 1982–85, NIVA conducted an extensive study of the mercury content of the bottom sediments of the lake. The study concluded that the sediments contained approximately 1.8 tonnes of mercury; half of which came from the Mesna cardboard factory. The remaining half was divided between the source areas in the Gjøvik-Hamar region. The results from the measurement of mercury in five rivers have been presented in Table 7. However, all of the tests were below the detection level, and have therefore not been discussed further.

4.3 Direct sources from treatment plants

4.3.1 PCBs and BDE-209

Reference is made to the report from Jordforsk (Snilsberg et al. 2005) for more detailed information on brominated flame retardants and other components. Selected samples from nine treatment plants in the region were further analyzed for PCBs, and the results have been presented in Table 8. The bottom of the table shows the estimated emissions based on annual rates of outflow from the plants. As the table shows, the results indicate that some 18 g ΣPCB_7 and some 46 g BDE-209 were released to Lake Mjøsa through outflow water from the nine treatment plants in question.

It must be pointed out that these calculations are based on limited numerical data, and that they only take into consideration regular outflow water from these plants as operated under normal conditions. Two selected samples from Lillehammer and Rambekk (Gjøvik) were also analyzed for levels in inflow water to the treatment plants. For BDE-209 the inflow water levels were 16 to 39 times higher than in the level in outflow water, while the corresponding levels for ΣPCB_7 were from 4 to 8. This means that under normal operating conditions without overflow from the treatment plants, the concentration level of PCBs and particularly BDE-209 will be significantly reduced in the treatment plants. Correspondingly, it appears reasonable to assume that the direct discharges of PCBs and BDE-209 to the lake can be expected to increase during flood periods if the inflow water is able to pass freely into Lake Mjøsa.

Table 8. Concentrations (ng/L) and discharges (g/year) of BDE-209 and PCBs through outflow water from nine selected treatment plants. Outflows from the treatment plants in litres per second are indicated in parenthesis in the left column. The estimates are uncertain (see the text).

Concentrations in outflow water (ng/L), 17–22/12 2004									
Treatment plant	**BDE-209**	PCB-28	PCB-52	PCB-101	PCB-118	PCB-138	PCB-153	PCB -180	**ΣPCB_7**
HIAS (222)	1.05	0.06	0.06	0.08	0.07	0.10	0.09	0.12	0.6
Nes (6)	1.67	0.04	0.05	0.08	0.05	0.10	0.11	0.09	0.5
Moelv (26)	1.31	0.06	0.06	0.05	0.04	0.06	0.07	0.06	0.4
Lillehammer (181)	1.92	0.19	0.24	0.20	0.15	0.20	0.20	0.18	1.4
Rambekk (139)	4.67	0.05	0.06	0.05	0.04	0.07	0.06	0.07	0.4
Breiskallen (78)	1.85	0.08	0.21	0.38	0.31	0.26	0.23	0.10	1.6
Kapp (5)	1.90	0.11	0.26	0.30	0.20	0.13	0.14	0.05	1.2
Skreia (11)	1.35	0.05	0.05	0.04	0.02	0.05	0.05	0.03	0.3
Lena (8)	0.75	0.04	0.04	0.04	0.03	0.05	0.04	0.04	0.3
Discharge (g/year)									
Treatment plant	**BDE-209**	PCB-28	PCB-52	PCB-101	PCB-118	PCB-138	PCB-153	PCB -180	**ΣPCB_7**
HIAS (222)	7.37	0.43	0.45	0.55	0.52	0.68	0.65	0.83	4.1
Nes (6)	0.33	0.01	0.01	0.02	0.01	0.02	0.02	0.02	0.1
Moelv (26)	1.08	0.05	0.05	0.04	0.03	0.05	0.06	0.05	0.3
Lillehammer (181)	10.94	1.10	1.38	1.13	0.83	1.12	1.14	1.01	7.7
Rambekk (139)	20.45	0.22	0.26	0.24	0.17	0.29	0.28	0.29	1.8
Breiskallen (78)	4.57	0.21	0.52	0.94	0.78	0.63	0.58	0.25	3.9
Kapp (5)	0.31	0.02	0.04	0.05	0.03	0.02	0.02	0.01	0.2
Skreia (11)	0.46	0.02	0.02	0.01	0.01	0.02	0.02	0.01	0.1
Lena (8)	0.20	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.1
Total treatment plants	**45.7**	2.1	2.7	3.0	2.4	2.8	2.8	2.5	**18.3**

4.3.2 Mercury from treatment plants

Two data sets have been given for mercury from treatment plants:

In the first data set (pre-project analyzed by NILU), one sample of outflow water from each of the nine treatment plants was analyzed for mercury, and the results have been presented in Table 9. Measured flows calculated using annual emissions from these plants are also presented in the same table. The data indicates that between 2 and 3 kg Hg is fed to Lake Mjøsa from treatment plants. The great level of uncertainty of these estimates may be seen from the fact that if one assumes that the one sample from Breiskallen is not representative of the plant, the level is reduced from 2–3 kg to 0.17 kg. One sample from Lillehammer and Rambekk (Gjøvik) each were also analyzed for levels in inflow water to the treatment plants. The sample from Rambekk was 20% higher than the level in the outflow water, while the corresponding value for Lillehammer was a little lower than for outflow water. Rambekk and Lillehammer have mercury input from fed quantities of external sludge (Rambekk: sludge from other treatment plants in Oppland; Lillehammer: septic sludge) and these sources are not registered in the tests taken of the inflow water. This explains how there can be higher values in the outflow water than in the inflow water. Rambekk has a mechanical/chemical treatment process, while Lillehammer has a biological/chemical treatment process, with removal of nitrogen. There is nothing tindicating that the treatment processes will lead to raised mercury levels (Paulsrud, Aquateam pers. comm.). Operational assistance (OA) for Oppland and Hedmark has some data on mercury in outflow water from several treatment plants in the region. They are labelled with an asterisk in Table 9. The median for Lillehammer and Rambekk has been used, and a single value from the HIAS organization has been used to calculate Hg emissions to Lake Mjøsa. The Lillehammer and Rambekk treatment plant represent about half of the emission volume to Lake Mjøsa from Oppland. HIAS represents about 90% of the emissions to Lake Mjøsa from Hedmark. The median for Rambekk is significantly higher than for the two other plants, and also in relation to the single value measured during the pre-project.

Table 9. Concentrations (ng/L) and sources (g/year) of Hg through outflow water from nine selected treatment plants. Emissions from the treatment plants in litres per second are indicated in parenthesis in the left column. The estimates are uncertain (see the text).

Treatment plant	Hg in inflow water (ng/L) (NILU pre-project)	Hg in outflow water (ng/L) (NILU pre-project)	Hg in outflow water (ng/L) (OA)	Hg in emissions g/year (NILU + OA)	Hg in sludge (µg/kg TS) (NILU pre-project)
HIAS (222)		6	7	42–49*	
Nes (6)		198		37	
Moelv (26)		17		14	
Rambekk (139)	8	6	150	26–657*	1151
Lillehammer (181)	<5	9	9	51	1934
Breiskallen (78)		903		2221	
Kapp (5)		<5		1	
Skreia (11)		<5		1	
Lena (8)		5		1	
Treatment plant (low–high) * OA data ** including Breiskallen				170–3000**	

Both sampling and analysis are a challenge for mercury in in/out flow water from treatment plants, and the results show that large series of samples are necessary in order to be able to exclude random errors in the sampling and/or analysis, which is clearly reflected in the estimate being between 0.17–3 kg. The same uncertainties regarding overflow as mentioned in chapter 4.3.1 also apply to mercury, and a total estimate of 0.17–3 kg from the treatment plants would merely be an indicative level. We strongly recommend quality assurance of this through further analyses.

There is also more data on mercury in sewage sludge from the municipalities around Lake Mjøsa for the period of 1989 up to the present. The average concentration in 2002 was from 0.5–2.2 mg/kg dry matter. Mercury concentrations from 1989 up to the present have shown a decreasing trend. This data can be found in the Norwegian Pollution Control Authority's pollution database. The sludge from Lillehammer is dewatered there, transported to Rambekk, mixed with other sludge and deposited in digesters. It is then dewatered, dried, pelleted and driven to the Dalborgmarka waste reception site. In the past, the sludge was driven to Nygard, and used as a top layer / finishing. Some sludge is used as a top layer / finishing at

Dalborgmarka. Some sludge is still simply deposited at Dalborgmarka. For the present, the sludge from Rambekk has been left at Dalborgmarka, and is not used for agricultural or green areas.

5. Development and Validation of a Model for Organic Contaminants in Lake Mjøsa

5.1 Background for the choice of model

The main goal of the development and application of a model has been to investigate whether observed levels of selected environmental contaminants in Lake Mjøsa can be explained based on current knowledge of existing sources. The model can then be useful in an initial attempt to understand and predict the environmental fate of selected contaminants in the lake. The model can thus provide important feedback in terms of detecting key knowledge gaps related to sources and the behaviour of pollution in the environment, and in the assessment of the effect of possible control measures in order to reduce the burden on the environment.

In developing the model, it is important to try to find the best balance between realism and data availability. Realism means a model that provides a satisfactory scientific and detailed description of the different processes that affect the contaminants' behaviour in the lake, and which can be useful for decision-makers in relation to the assessment of control measures. Data availability here means the necessary information for a quantitative description of different processes in the model, and the necessary information from past monitoring efforts in order to be able to evaluate (validate) the model's results with observations. In general, one can assert that detailed, realistic models are very demanding in terms of input data, while knowledge of the processes and parameters that guide the behaviour of the contaminants in question in Lake Mjøsa is still very limited. The model developed is therefore relatively simple in order to minimize the risk of over-parameterization, in terms of available information

As the Norwegian Pollution Control Authority has also considered it important to be able to evaluate scenarios regarding the future environmental burden on Lake Mjøsa in this project, we have decided to develop a dynamic model for the lake. By dynamic, we mean that the model has been developed to be able to assess changes to the environmental burden over time, as a function of temporal changes in emissions.

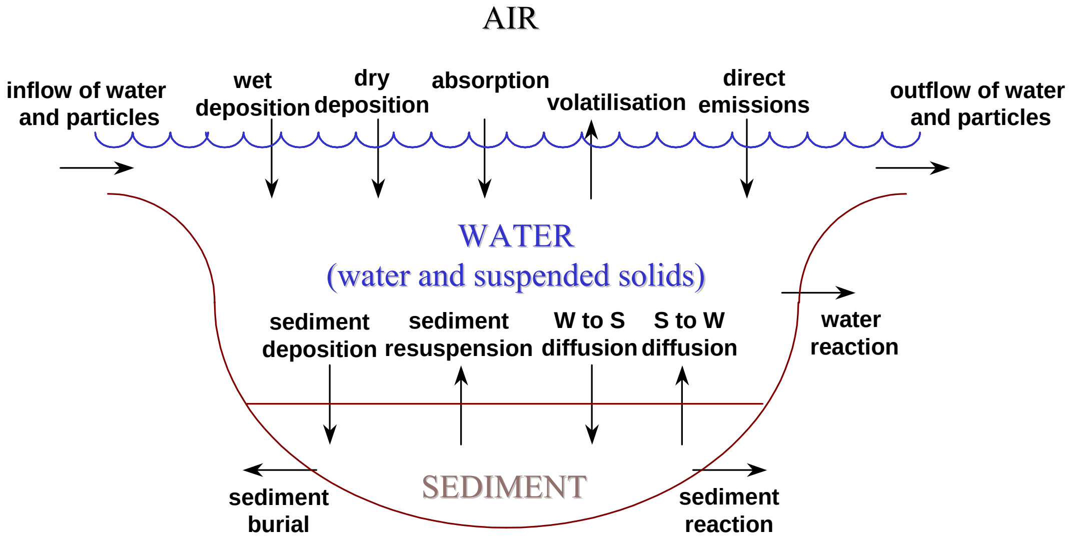


Figure 19. Key processes in Mjøsa-POP (according to Mackay, 2001). The key processes in the model are identical with the QWASI model (Mackay et al., 1983), while the process descriptions are deviant in several cases (Breivik et al., 2003).

To a great extent, the model is based on the QWASI model by Mackay et al. (1983). See Figure 19. The QWASI model (Quantitative Water Air Sediment Interaction Model) is a so-called fugacity-based steady-state non-equilibrium multimedia model for lakes (Mackay, 2001). It focuses on descriptions of exchange processes between water and sediments, and between water and air. These are relevant processes to include, as it has long been known that contaminants can leak out from sediments (Larsson, 1985), and that evaporation of contaminants from lakes to air can occur after following emission reductions (Jeremiason et al., 1994).

The QWASI model is considered a robust model tool, as it has been tested and evaluated for many different contaminants and environments (Mackay, 2001). The QWASI model further represents the core of newer, more complex non-steady state models (see e.g. Warren et al., 2002), which have been developed and evaluated for organic contaminants in the Baltic Sea, among other places (Wania et al., 2000; Breivik & Wania, 2002), and the inner Oslo fjord (Breivik et al., 2004b). The model for Lake Mjøsa represents a modified version of the model that has been developed for the inner Oslo fjord (Breivik et al., 2003). However, the model that has been developed for this project does not have any spatial resolution, which means that it attempts to describe the conditions for Lake Mjøsa as a whole. However, the process descriptions are identical with the model for the inner Oslo fjord. See Breivik et al. (2003) for a detailed description of all equations and process descriptions.

5.2 Input data

The model needs three different types of input data (parameters) to describe the cycling and environmental fate of the contaminants. They are (*i*) parameters that describe the propoerties of contaminants – see 5.2.1, (*ii*) parameters that describe the properties of Lake Mjøsa – see 5.2.2 and (*iii*) information about emissions and discharges of the components in question – see 5.2.3.

5.2.1 Physical-chemical data and decomposition speeds

The input data that describe the environmental contaminants can be divided into physical-chemical property data, and degradation rates. Table 10 shows the selected values of key PCB compounds.

Table 10. Physical-chemical data and PCB half-lifes (at 25˚C)

	PCB-28	PCB-52	PCB-101	PCB-118	PCB-138	PCB-153	PCB-180
Molecular weight [g mol^{-1}] A)	257.5	292.0	326.4	326.4	360.9	360.9	395.3
log K_{OW} A)	5.66	5.91	6.33	6.69	7.21	6.87	7.16
dU_{OW} [kJ/mol] A)	-26.3	-27.3	-23.8	-28.5	-25.0	-31.1	-29.1
log K_{AW} A)	-2.18	-2.31	-2.40	-2.67	-2.44	-2.57	-3.00
dU_{AW} [kJ/mol] A)	52.3	54.1	59.7	60.5	61.3	62.8	63.6
$t_{1/2}$ water2) [1000t] B)	5.5	10	31	46	46	55	55
$t_{1/2}$ sediment2) [1000t] B)	17	55	55	100	100	170	170
Activation energy [kJ/mol] C)	30	30	30	30	30	30	30

A) Li et al., (2003) B) Based on information in Wania and Daly (2002) supplemented with values from Breivik et al., (2004b). C) Wania and Daly (2002).

K_{OW} and K_{AW} offer important information used to calculate the phase distribution of contaminants between air, water and organic matter (particles in air, water and sediments), while the half-lifes given are an estimate of the decomposition in the environment. The temperature dependency for phase distribution is described using enthalpies of phase transfer (dU_{OW} and dU_{AW}), while the activation energy says something about the presumed temperature dependency for degradation processes. The input data that describes degradation rates in the environment are considered uncertain (Gouin et al., 2004), while the physical-chemical input data are considered fairly reliable, as they are based on a careful analysis of all the empirical data that are available (Li et al. 2003).

5.2.2 Site-specific data

A brief description of the input data used to describe Lake Mjøsa as a system follows. Table 11 provides static input parameters that are heavily based on previous studies by NIVA, as part of the Lake Mjøsa action (1976–1981) and regular monitoring programs (see e.g. Holtan et al., 1979; Kjellberg et al., 1982/1986; Kjellberg, 1994; Nashoug, 1999). The Norwegian Meteorological Institute has also provided the project with meteorological data (Elin Lundstad, pers. comm.). Some of the input data is unknown or is not determined for the lake. These input data have therefore been based on estimates from different experts at NIVA, NILU, and from the literature. It must therefore be pointed out that there is significant uncertainty about some of the input data. Further investigations are necessary in order to achieve a better characterization of them in order to achieve more reliable model results.

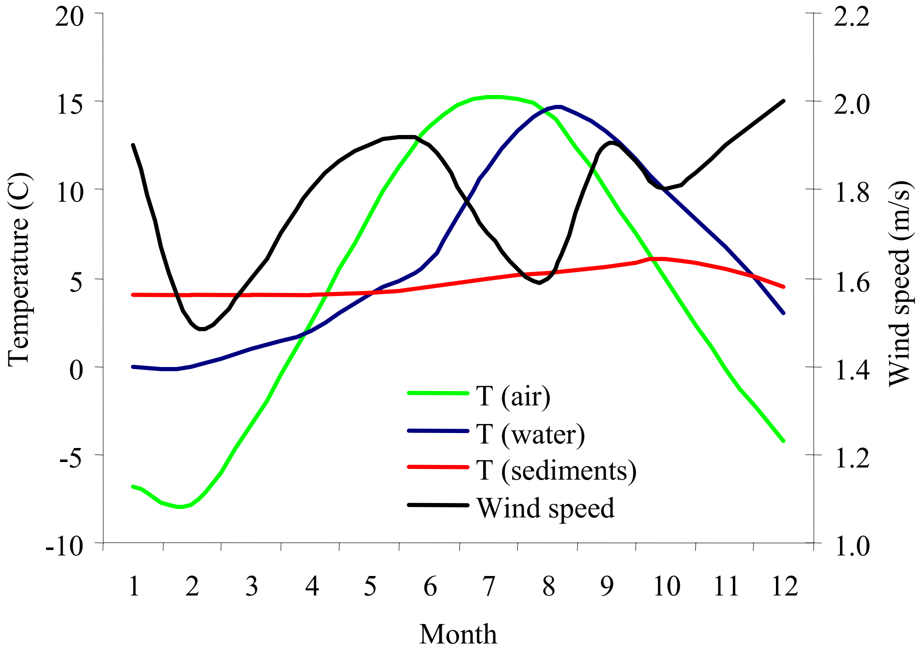


Figure 20. Input data with seasonal variation (based on long time series from NIVA and the Norwegian Metorological Institute).

Some input data has been provided with seasonal variation (Figure 20). Temperature variations determine the temporal variability in phase distribution and degradation rates (Breivik et al. 2003), while wind speed is part of the description of gas exchange of contaminants between the atmosphere and water (Mackay and Yuen, 1983). Air temperatures and wind speeds are based on monthly averages for Kise in Hedmark for the period of 1951–2004 (Elin Lundstad, pers. comm.). Estimating seasonal variations in the temperature of the lake has been a major challenge when viewing Lake Mjøsa as a uniform system without spatial resolution (see e.g. Kjellberg et al., 1982). To operate with a simple parameterization, we have therefore decided to use the temperature measured at defined depths in Skreia in 1989–2003 (Gösta Kjellberg, NIVA, pers. comm.). The water temperature in the model (Figure 20) reflects the average water temperature at 10 m depth during the period, while sediment temperatures in the model refer to water temperatures at 50 m depth.

Table 11. Site-specific input data. Parameters in italics *indicate input data that is considered highly uncertain.*

Dimensions		***Density***	
Surface area of Lake Mjøsa [m^2] A)	$3.62 \cdot 10^{8}$	Octanol [kg m^{-3}] K)	820
Volume [m^3] A)	$5.62 \cdot 10^{10}$	Inorganic solids [kg m^{-3}] K)	2400
Active sediment depth [m] B)	0.04	Organic matter [kg m^{-3}] K)	1000
Hydrology		***Other sediment-related parameters***	
Annual precipitation volume [mm/year] C)	585	Sedimentation rate [mm year^{-1}] L)	*1*
Inflow of water to Lake Mjøsa [m^3 sec^{-1}] D)	323	Fraction buried in deeper sediment [-] M)	*0.75*
Outflow of water from Lake Mjøsa [m^3 sec^{-1}] D)	320	***Particle/solid concentration***	
Particular fraction of organic matter (OM) or organic carbon (POC)		Inflowing water [mg L^{-1}] E)	*3*
Inflowing water [g OM g^{-1} particles] E)	*0.45*	Lake Mjøsa [mg L^{-1}] N)	0.6
Lake Mjøsa [g OM g^{-1} particles] F)	*0.53*	Air [µg m^{-3}] O)	6
Air [g POC g^{-1} particles] G)	*0.15*	Volume fraction of solids in sediment [m^3 m^{-3}] M)	0.63
Sediment [g POC g^{-1} particles] H)	0.03	***Diffusion parameters and mass transfer coefficients (MTC)***	
Resuspended sediment [g POC g^{-1} particles] I)	0.03	Molecular diffusivity in water [m^2 hour^{-1}] M)	*$4 \cdot 10^{-6}$*
Other atmospheric parameters		Diffusivity in sediments (bioturbation) [m^2 hour^{-1}] J)	*$1 \cdot 10^{-10}$*
Particulate dry deposition velocity [m hour^{-1}] J)	1.03	MTC for water-sed. diffus. (water side) [m hour^{-1}] M)	*0.01*
Scavenging ratio [-] J)	*68000*	MTC for gas exchange between air and water	See text

A) Kjellberg (1994) B) Assumed value (Gösta Kjellberg, NIVA, pers. comm.) C) Data for Kise, 1951–2004 (Elin Lundstad, pers. comm.) D) Assumed somewhat higher than outflow (Gösta Kjellberg, NIVA, pers. comm.) E) Calculated based on reported sources of inorganic and organic particular material in Holtan et al. (1979) F) Holtan et al. (1979) G) Karl Espen Yttri, NILU, pers. comm. based on values in Aas et al. (2002; 2003; 2004) H) Aagard et al. (1976) and Fjeld et al. (2004) under the assumption that TOC ≈ POC I) Assumed equal to sediment. J) Wania et al. (2000) K) Daly and Wania (2002) L) Based on information in Berge et al. (1973); Aagard et al., (1976); Holtan et al. (1979) M) According to Mackay (2001) N) Based on data from Holtan et al. (1979) and Gösta Kjellberg, NIVA (pers. comm.) O) Leiv Håvard Slørdal, NILU, pers. comm./ Aas et al. (2002; 2003; 2004).

5.2.3 Sources and Discharge

An important part of the project has been to achieve a better quantiative understanding of the sources and pathways of selected environmental contaminants to Lake Mjøsa, focusing on the atmospheric burden. This is discussed in chapter 3. Problems with PCB detection levels in water samples from the rivers (Table 7) have prevented the quantification of sources via rivers, based on measurements. As a temporary solution to the lack of reliable measurements, we have decided to calculate the inflow of contaminants from rivers by scaling the results for the average of the four water samples from Kise (Table 5), based on the results from the relevant passive water samples taken from the rivers (Table 6). The conversion factors are presented in Table 12.

Table 12. Average concentration in water measured in Kise (pg/L), conversion factors based on SPMD measurements, and calculated concentration in selected rivers.

PCB	Measured in Kise	Conversion factor (SPMD)			Calculated concentration		
		Låg/Kis	Mes/Kis	Hun/Kis	Låg	Mes	Hun
	pg/L	–	–	–	pg/L	pg/L	pg/L
PCB-28	2.2	0.41	0.37	4.71	0.9	0.8	10.1
PCB-52	2.5	0.29	0.50	3.24	0.7	1.2	8.1
PCB-101	0.7	0.32	0.78	2.95	0.2	0.6	2.1
PCB-118	0.3	0.34	1.04	3.37	0.1	0.3	1.0
PCB-138	0.3	0.41	0.92	3.18	0.1	0.3	1.1
PCB-153	0.4	0.41	0.84	2.13	0.2	0.3	0.8
PCB-180	0.1	0.52	1.30	3.71	0.1	0.1	0.3

Based on this data, a table has been drawn up to quantify sources via air, rivers and direct sources (treatment plants) in the model in Table 13. The concentration in rivers indicates an averaged concentration in relation to average water flow, based on the values calculated in Table 12 and average water flow in the Lågen, Mesna and Hunnselva (Kjelberg et al. 1982). However, the Lågen, Mesna and Hunnselva represent 90% of the average water flow from the 15 largest rivers that run out into Lake Mjøsa (Kjelberg et al. 1982).

Table 13. Input data used to describe sources of contaminants to Lake Mjøsa in the model.

Compound	Air concentration (pg/m^3) [A]	Concentrations in rivers (pg/L) [B]	Direct sources (g/year) [C]
PCB-28	1.7	1.0	2.1
PCB-52	1.5	0.9	2.7
PCB-101	0.9	0.3	3.0
PCB-118	0.4	0.1	2.4
PCB-138	0.5	0.2	2.8
PCB-153	0.7	0.2	2.8
PCB-180	0.2	0.1	2.5
ΣPCB_7	6.0	2.7	18.3

[A] As described earlier – see section 4.1.1. [B] See text [C] Based on data from nine treatment plants. See section 4.3.

It must be pointed out that the conversion factors introduce significant uncertainty to the calculation of an average concentration for rivers that run out into Lake Mjøsa. There is further a risk of a water sample leading to underestimation of the actual input from the rivers. This can be justified by the possibility of some particular matter being transported along the riverbed. PCBs have an increasing affinity for organic matter in particles with an increasing level of chlorination. If there is significant transport of PCBs in particles along the riverbed, this can create an additional contribution in excess of what the water samples can capture. To test this hypothesis, we tried to estimate the transport of PCBs in particles, based on sediment concentrations measured in the Lågen from stations Låg-1, 2, 3 (Fjeld et al. 2004) and compare them with the data expected for the Lågen (see Table 12). Data from station Låg-4 (Svartevjua outside Jørstadmoen) has not been included, as this sample was not considered representative for the main course by Fjeld et al. (2004). The PCB concentration in sediment was first normalized in relation to organic content in the sediment samples from the riverbed. The average was then calculated and corrected against the average inflow of particular organic matter from the Lågen (Holtan et al. 1979) and the average water flow for the Lågen (Kjellberg et al. 1982). The results are shown in Table 14.

Table 14. Calculated concentration of PCBs in the Lågen, based on conversion via SPMD samples (Tables 10 and 11), and transport of particular organic matter alone (as described above).

Compound	Concentration in the Lågen based on conversion via SPMD (pg/L)	Concentration in the Lågen based on particle transport (pg/L)
PCB-28	0.9	0.9
PCB-52	0.7	2.2
PCB-101	0.2	4.5
PCB-118	0.1	3.6
PCB-138	0.1	4.5
PCB-153	0.2	4.8
PCB-180	0.1	2.0
ΣPCB_7	2.3	22.4

As can be read in Table 14, the results show that the alternative calculation generally yields far higher PCB concentrations. The concentration is 10 times greater for ΣPCB_7 if the concentration in the Lågen is calculated based on sediment concentration. It is also clear that the relative deviation between the two calculations increases with the chlorination level, which can be explained by the increasing particle affinity from PCB-28 to PCB-180. However, there is a risk of overestimation of the actual particle transport in sediment-based concentrations, because the inflow of organic matter is probably significantly lower than in 1973–1976 (Holtan et al. 1979). This example illustrates the relationship between different environmental problems like eutrophication and organic contaminants (Skei et al. 2000). It thus appears less probable that the concentration in the Lågen (as calculated based on particles) is actually 3–4 times greater than that measured outside Kise (Tables 5 and 12), which the SPMD measurements (Table 6) also show is incorrect. On the whole we have therefore chosen to use the calculated concentrations given in Table 13 of the model. We should still remember that sources via rivers are both uncertain and may be significantly underestimated. To quantify the relative significance of sources of contaminants from rivers with a greater degree of certainty, follow-up investigations are recommended.

5.3 Evaluation of the model against observations

The levels of environmental contaminants observed in the lake today must be understood from the perspective of historical and current sources (as the contaminants have a long life in the environment). It has therefore been necessary to reconstruct the history of pollution in the model over a time period that reflects the life of the relevant compounds in the lake. It is difficult to do this in satisfactory fashion, as the relevant pollution history for Lake Mjøsa is unknown. A common technique for shedding light on past sources of environmental contaminants is to analyze dated sediment cores (e.g. Christensen & Lo, 1986; Koneczny et al. 1994; Bruckmeier et al., 1997; Gevao et al. 1997). However, such analyses have not been conducted in Lake Mjøsa. To avoid unrealistic assumptions regarding constant emissions, the emissions for 2004 (Table 13) were scaled back in time in relation to national data on PCBs in use in 1980, 1995 and 2002 (Norwegian Pollution Control Authority 2005). Between 1995 and 2002, the amount of PCBs in use was reduced by about 5% per year. A corresponding reduction is assumed for 2002–2004.

It has not been possible to model BDE-209 in this project, due to inadequate data on sources via rivers and lack of data for evaluating the model against observations.

5.3.1 Evaluation of PCBs

5.3.1.1 Water

Measurements of PCBs in water outside Kise are an important reference in order to be able to assess whether the model complies with observations. The results of the measurements and the model are compared in Figure 21. The results clearly show that the model underestimates observed concentrations (Figure 21A), while the relative distribution (composition in %) has been satisfactorily reproduced (Figure 21B). For ΣPCB_7 the modelled concentration is 22% of the observed average.

Underestimation of observed concentrations may be due to three factors:

1) Underestimation of PCB inputs to the lake.
2) An error in the model (in descriptions of different processes).
3) The sample from Kise not being representative of the general level in the water masses.

Based on current knowledge, it is not possible to say with certainty whether the underestimation is the result of the inputs of PCBs being too low (1) or an error in the model (2). It is natural to assume that inadequate representativity (3) is not the primary cause of deviation between observations and the model, based on results from the passive water samples (see Table 6), even though one cannot disregard the fact that precisely these samples have been influenced by a local source.

However, what can be said with certainty is that the sources necessarily carry a bias towards underestimation. This has been discussed earlier in relation to treatment plants (where sources via overflow have not been quantified), for sources via rivers (where the calculations probably do not take satisfactory consideration of transport of particles along the riverbed) and air (where the samples are from the cold season, with presumably lower levels). One cannot simply disregard that other land-based sources of PCBs to the lake can make a significant contribution to the general burden level of PCBs in the lake. Possible candidates can be overflow from treatment plants during flood periods and runoff from soil (see chapter 4.3), landfills, industrial areas, cities and towns. Neither can increased discharges be overlooked during snow melts (Daly & Wania, 2004). Road traffic has also been mentioned as a possible source of PCBs. However, the passive air samplers that have been located close to the E6 motorway outside city areas (Vingrom, Minnesund) indicate that road traffic does not make a significant contribution to atmospheric sources. In total, the sources have been quantified as small (see Tables 13 and 16) when compared with the PCB volumes that have been used in Norway (Norwegian Pollution Control Authority 2005) and the amount of PCB contained in various products. For example, capacitors in old fluorescent tube components may contain 30–70 g pure PCBs. This clearly illustrates that if even small volumes of waste containing PCBs end up in Lake Mjøsa and leak out to the surroundings, this can make a significant contribution to the environmental burden in the lake.

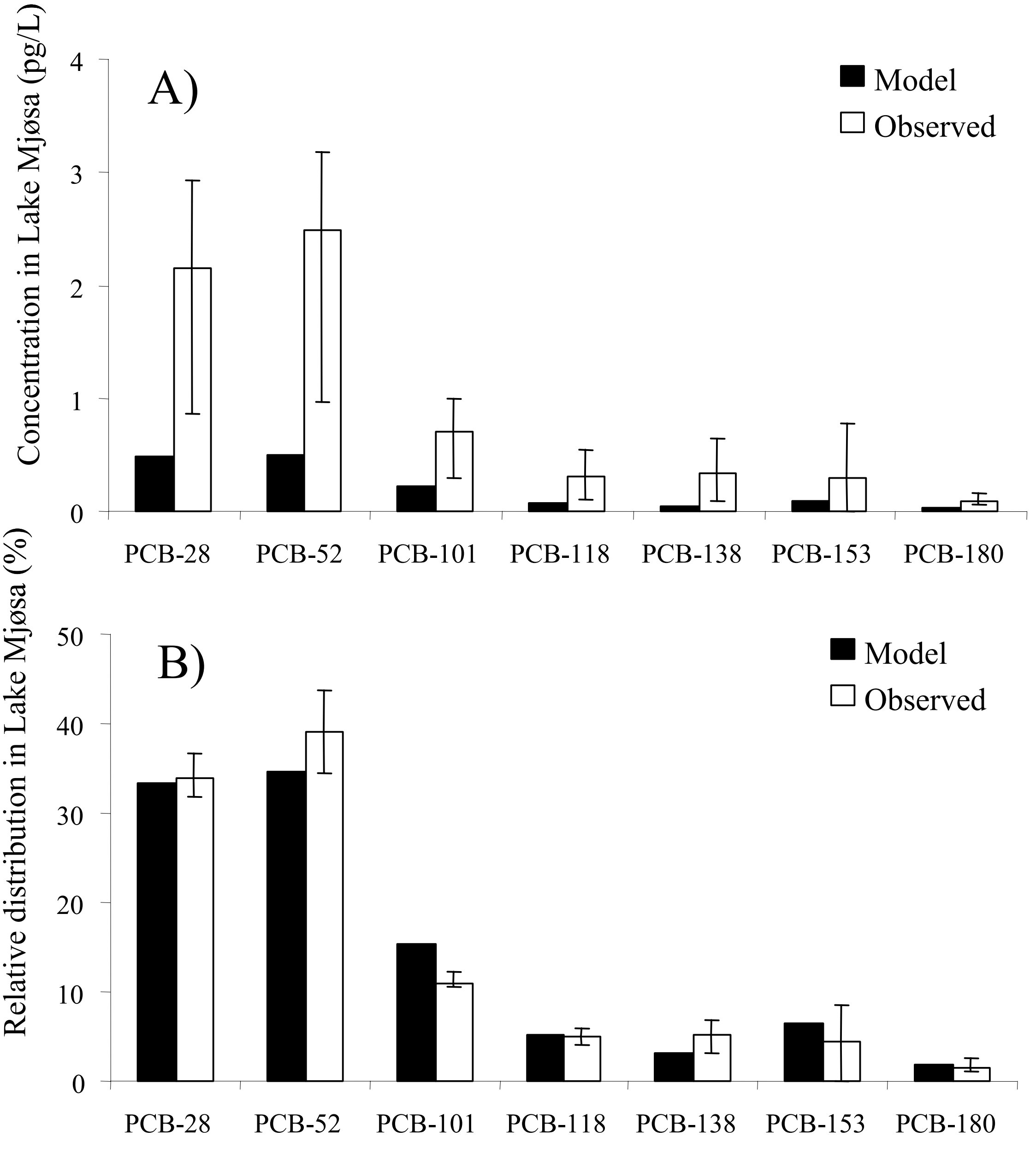


Figure 21. Comparison of observed (average) and modelled total concentration of various PCB compounds in water from Lake Mjøsa (Kise), autumn 2004. A) Absolute concentrations. B) Relative distribution of different compounds. Vertical lines for observed concentrations indicate maximum and minimum values.

Further source investigations appear to be necessary in order to achieve an improved characterization of the sources that control the PCB levels in Lake Mjøsa. A possible strategy is a more comprehensive study to evaluate spatial variation of contaminants in stationary species in the lake or in sediments in relation to potential emission points / source areas. The

PCB measurements in air were further conducted from September 2004 up to and including January 2005. As mentioned earlier, the results from this and other studies indicate that the PCB levels are generally higher during the warm season than in the cold season (Haugen et al. 1999). The actual atmospheric inputs may thus be somewhat underestimated in the model, without this being able to explain the entire deviation between model and observations.

The deviation between model and observations can also be due to errors in the model. Significant uncertainty is linked to the description and quantification of exchange processes between water and sediments. As mentioned earlier, the PCB levels in Lake Mjøsa are due to both current emissions and past discharges. There is a risk of the model overestimating different loss processes for PCBs in the lake, which will imply an underestimation of the concentrations observed. Further, the Lake Mjøsa pollution history and leakage of PCBs from sediments may be underestimated in the model, which will also imply that the model underestimates observed concentrations. However, it must be pointed out that the model is able to reproduce the relative distribution of PCBs in water, which provides an indication that the PCB properties have been described adequately in the model.

5.3.1.2 Sediments

Issues related to representativity make it very challenging to compare results from the model with observations related to sediments. This is because sediments are expected to show far greater spatial variability in terms of PCB levels, compared with levels in water. This particularly applies to locations where direct emissions by air or water to Lake Mjøsa are taking place or have taken place. As yet, no investigations have been conducted with a view to achieving a representative picture of the general environmental burden on the sediments in Lake Mjøsa, corresponding to previous investigations of the Oslo fjord (Konieczny et al. 1994).

Table 15. Results from some past investigations of ΣPCB_7 in sediments in Lake Mjøsa (in µg ΣPCB_7/kg dry weight).

Location	Year	ΣPCB_7	Reference
Totenvika	1990	0.5	Kjellberg, 1991
Tangenvika	1990	2.3	Kjellberg, 1991
Åkersvika	1990	1.2–326	Kjellberg, 1991
Mjøsa[A]	1996/1997	1–30	Rognerud et al. 1997
Mjøsa[B]	1996	5.4–11	Kjellberg pers. comm.
Hamar (n=30)	1999	0.2–121	Kjellberg and Løvik (2000)
Lågen (n=4)	2004	0.99–20.01	Fjeld et al. 2004
Vorma (n=2)	2004	0.87–1.13	Fjeld et al. 2004
Model	2004	0.24	

[A] The samples were taken in Lillehammer, Gjøvik, Brummundal, Hamar and Totenvika. [B] Samples from Furnesfjorden (60 m) and Hamarfjorden (160 m). Unpublished data from Gösta Kjellberg, NIVA.

For example, Kjellberg and Løvik (2000) investigated concentrations of surface sediments (0–2 cm) from 30 locations near NSB's workshop area in Hamar and found significant spatial variation in 1999 (0.2–121 µg ΣPCB_7/kg dry weight). However, the purpose of their investigation was to assess local levels near a known pollution source. Comparison of model results from local hot spots with modelled concentrations that seek to provide data on the Lake Mjøsa levels as a whole must therefore assess observations based on representativity.

The results from the model yield an average concentration of PCBs in sediments of 0.24 µg ΣPCB_7/kg dry weight in 2004, while the measurements generally indicate higher levels (Table 15). It is worthwhile to emphasise that the ΣPCB_7 levels that have been measured in past studies and which do not have a bias towards locations with high levels of contaminants still appear to lie within the Norwegian Pollution Control Authority's condition category I (*insignificant – low pollution*), while the PCB levels in predatory fish are still over the limit value for dietary restrictions.

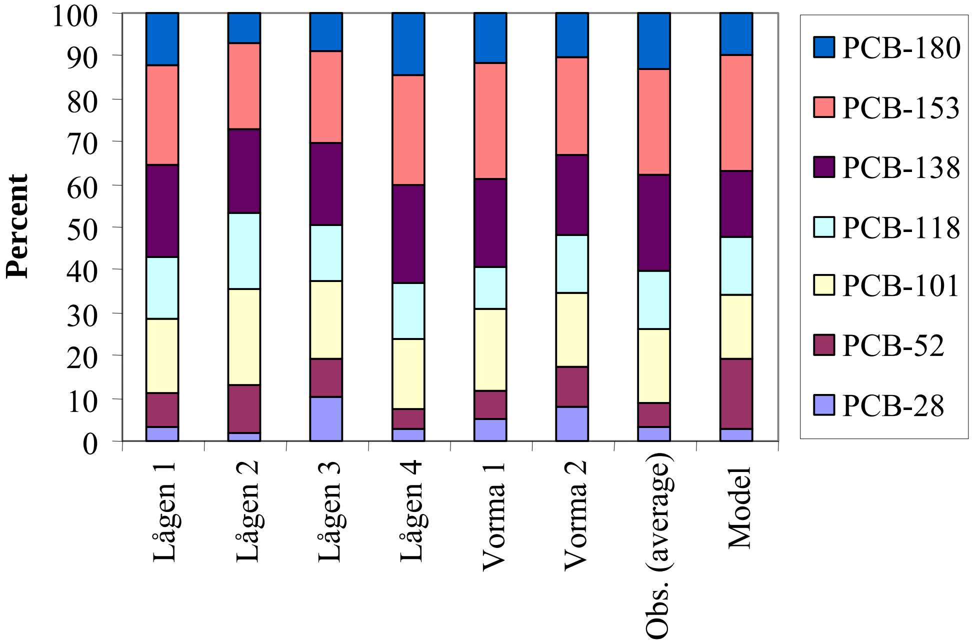


Figure 22. Relative distribution of different congenes in individual sediment samples from the Lågen and the Vorma (Fjeld et al. 2004), observed average for these samples and model predictions.

A comparison of relative distribution in sediments from 2004 is shown in Figure 22. Again it looks like the model is fairly close to the observed relative distribution in sediments.

6. Application of the Model for PCBs

6.1 Overview of PCBs in Lake Mjøsa, 2004

The evaluation of the model showed that the model underestimates concentrations observed in water and sediment, while the relative distribution of PCBs was reproduced satisfactorily. It also noted that there are few representative observations that the model can be compared with. This implies that it is difficult to have confidence in the absolute results generated by the model based on current input data (which guides the process descriptions). However, the model may contribute to providing a simplified – albeit uncertain – picture as a whole of the behaviour and environmental fate of PCBs for 2004. It may also be of significant value to future investigations that certain output data from the model is reported, so that new results can be compared with results from this examination and improve the knowledge basis.

Table 16. Mass balance of PCBs in Lake Mjøsa, 2004

		28	52	101	118	138	153	180	ΣPCB$_7$
Sources	**Total (g)**	**51**	**50**	**31**	**16**	**18**	**25**	**9**	**200**
	Atmosphere (%)	75	77	82	77	76	82	67	77
	Discharge (%)	4	6	10	15	16	11	27	9
	Rivers (%)	21	18	9	8	8	7	6	14
Loss processes	**Total (g)**	**61**	**82**	**60**	**35**	**40**	**54**	**20**	**352**
	Evaporation (%)	28	16	7	2	1	2	<1	10
	Degradation in water (%)	23	10	2	1	<1	1	<1	7
	Degradation in sediment (%)	35	44	56	48	49	36	36	44
	Outflow (%)	8	6	4	2	1	2	2	4
	Burial (%)	6	24	31	47	49	60	61	35
Sources / loss (g/g)		0.83	0.60	0.51	0.46	0.45	0.46	0.46	0.57
Volume	**Total (kg)**	**0.17**	**0.80**	**0.73**	**0.65**	**0.77**	**1.31**	**0.48**	**4.91**
	Water (%)	16	4	2	<1	<1	<1	<1	2
	Sediment (%)	84	96	98	>99	>99	>99	>99	98

The model indicates that the quantified sources are dominated by atmospheric deposition (77%), followed by rivers (14%) and direct discharges (9%). As mentioned earlier, the sources to Lake Mjøsa (and especially direct discharges) may have been underestimated.

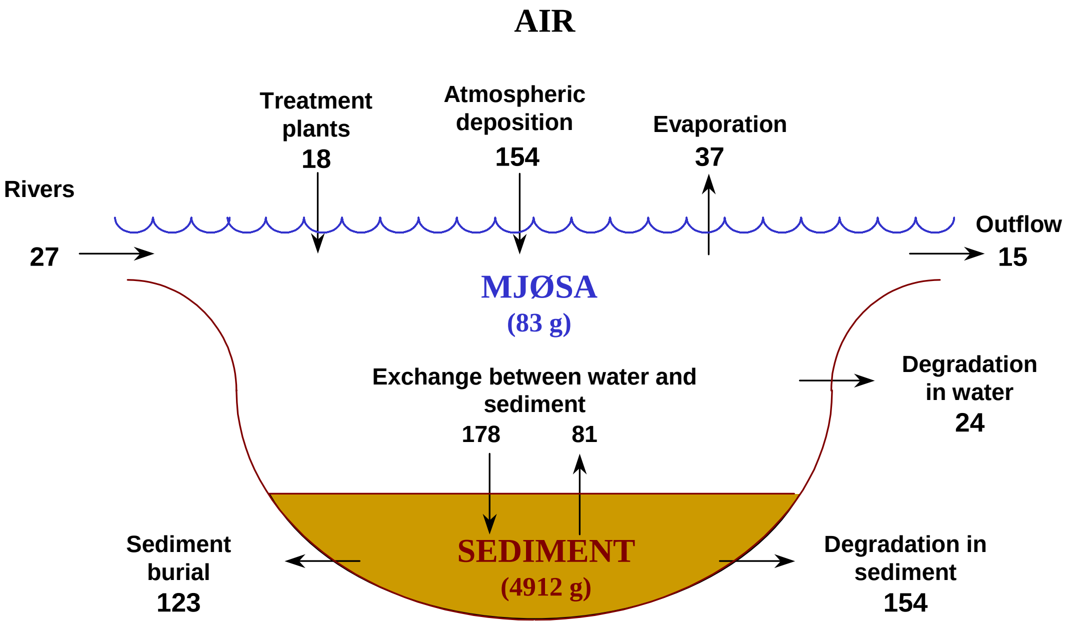


Figure 23. ΣPCB₇ mass balance calculated for Lake Mjøsa, 2004 (g per year). The amounts of ΣPCB₇ in water and surface sediment are also given in parenthesis.

PCBs disappear from Lake Mjøsa through five different processes: evaporation from the lake (10%), decomposition in water (7%) and sediments (44%), outflow via the Vorma (4%) and burial in deeper sediments (35%). The results further indicate that the total loss (in g) is almost twice as great as the input in 2004, which indicates the influence of former discharges vs. current sources on present levels in Lake Mjøsa.

The total amount of ΣPCB_7 in Lake Mjøsa (water plus surface sediment from 0–4 cm) has been calculated as almost 5 kg at the end of 2004. By way of comparison, Kjellberg and Løvik (2000) calculated the total volume of ΣPCB_7 in the surface sediment (0–2 cm) in Båthavnsvika near Hamar to be about 2.3 g ΣPCB_7. However, this does not mean that the local effects of the elevated levels in Båthavnsvika (or other places with heavily contaminated sediment) may be simply neglected.

6.2 Hypothetical scenario for future development

As the data for assessing the model's reliability is very limited, and the model turns out to underestimate concentrations observed, the model should not be used to assess future absolute concentration levels. However, the assessment indicates that the model is able to reproduce the relative composition of the lake adequately. Taking into consideration the significant uncertainties that have been described, the model can thus be used to attempt to shed light on relative changes in the future, given a defined scenario.

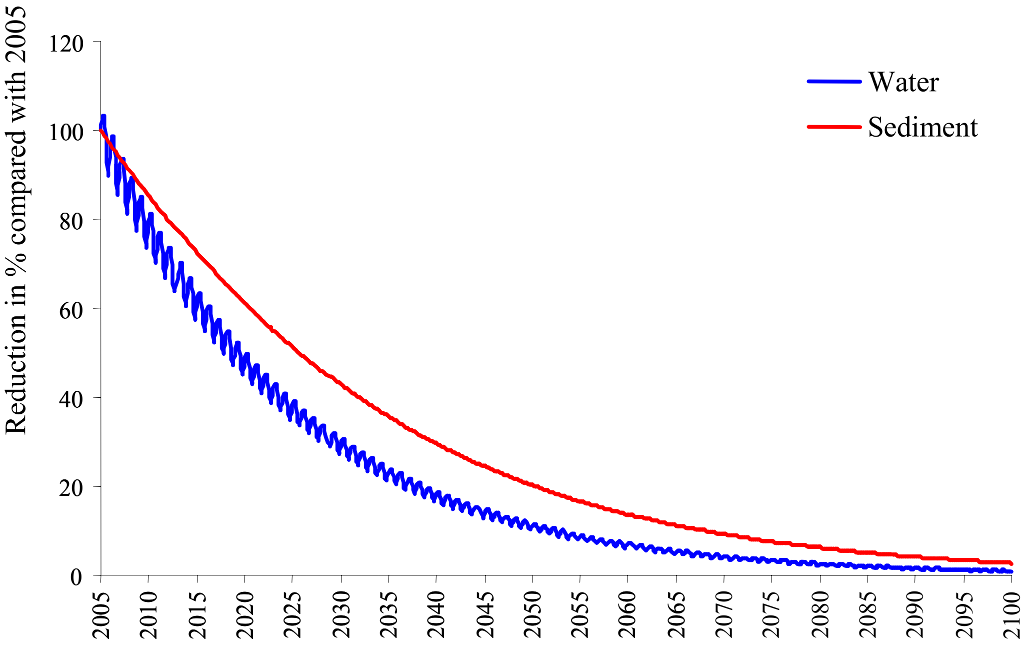


Figure 24. Estimated reduction in concentrations in water and sediment for ΣPCB$_7$ (in %) with a hypothetical 5% reduction of input per year from 2005 to 2100.

An example of one such scenario has been presented in Figure 24, where the sources of PCBs are reduced at a rate of 5% per year. With this scenario, the concentration in water would be reduced to 50% by 2019 (the year after the sources are reduced correspondingly) compared with the current level, while the concentration in sediment will be reduced to 50% by 2025. It is worth remarking that the sediment levels being halved would occur a mere 6 years after the corresponding reduction in water. It must finally be pointed out that this scenario outlines a hypothetical development, and is only intended to illustrate the potential of the modelling tool if a more accurate source characterization and systemic understanding (the latter through model improvement) are possible in the future. There is still no doubt that there will be PCBs in Lake Mjøsa for many years to come. An improved systemic understanding and source characterization may yield more reliable information on the time it will take, given the scenario defined for control measures.

7. Conclusions and Summary

7.1 PCBs

Overall, the results show that the PCB source pattern to Lake Mjøsa is complex, and that there may be significant sources of PCBs that have not been identified yet. However, the results from this project indicate that the atmosphere is an important source of PCBs to Lake Mjøsa, compared with an initial calculation of sources via rivers (based on calculated concentrations in water) and treatment plants (under normal operating conditions).

The PCB levels in the air around Lake Mjøsa are fairly typical for the background burden in Norway. The PCB levels in air over time are somewhat higher in the cities of Hamar and Gjøvik, which indicates that there may be a certain variability in terms of atmospheric sources around Lake Mjøsa, caused by local emissions to air in the region. By way of comparison, the PCB levels in Oslo are 10–50 times higher than the general atmospheric burden around Lake Mjøsa. Increased PCB levels in air coincide with transport episodes from the areas outside the Lake Mjøsa region. The Oslo region, the British Isles and Eastern Europe / Russia stand out as probable contributors. Other model calculations of long-range transport of PCBs to Norway indicate that the atmospheric deposition of PCBs far exceeds our national emissions. Investigations conducted under the Geneva Convention (CLRTAP) indicate that the long-range transported amount exceeded national emissions by a factor of 25 in 2000 (Mantseva et al., 2004). It is important to note that the estimate for Norway as a whole is uncertain, and that this factor can be expected to vary greatly for different parts of the country (in particular, a reduction to this factor is expected in places close to national source areas for PCB emissions to air). Proximity to the Oslo region may e.g. contribute to a significant reduction in this factor for Lake Mjøsa.

The results further indicate that land-based sources around Lake Mjøsa have not been adequately characterized and quantified. Overflows from treatment plants stand out as *one* probable source of increased PCB sources to Lake Mjøsa during flood periods, beyond that which has been calculated in this report. The transport of PCBs on particles along riverbeds is another source that may provide additional inputs, beyond the provisional estimate for sources via rivers.

The model that has been developed to shed light on quantitative relationships between sources and levels in the lake underestimates observed PCB concentrations. However, the relative distribution of different compounds appears to have been reproduced adequately. The results show a complex picture, where the levels in Lake Mjøsa can be explained on the basis of existing and historic sources. The model (which deals with both) can explain about one-fourth of the observed ΣPCB_7 levels, based on water samples collected outside Kise. The deviation between observed concentrations is primarily expected to be due to two factors: 1) Underestimation of PCB sources to the lake. 2) Errors and faults with the model. Current knowledge makes it difficult to determine which of these two factors is most important.

Uncertainty associated with the current empirical data makes it a challenge to issue specific recommendations regarding control measures that one can categorically say will lead to a significant reduction to the PCB burden in Lake Mjøsa. The results that have been presented and discussed are still useful for identification of central knowledge gaps, so that the basis on which decisions are made may be improved.

The atmosphere still stands out as a dominant source of current inputs that have been quantified, and further efforts to reduce PCB emissions to air may help reduce levels in the lake. However, the results indicate that episodes of increased burdens are mainly due to long-range transport from areas outside the Lake Mjøsa region. Further measures to reduce emissions to air should therefore be assessed in relation to national plans for phasing out PCBs, as well as through further follow-up of international agreements.

The results also indicate that other land-based sources of PCBs may have been underestimated. Local measures should therefore initially focus on achieving improved characterization of local land-based sources. Neither can one exclude sources within the lake *(leakage of PCBs from highly contaminated sediment or waste containing PCBs that may have come to rest in the lake)* having made a significant contribution to the general burden level in Lake Mjøsa. As sediment represents an important reservoir of PCBs, further examinations should be conducted to improve the systemic understanding of processes that are key to the exchange of PCBs between water and sediment. Any future studies should regardless focus on Lake Mjøsa as a whole, as the dietary advice applies to the whole lake.

7.2 PBDEs

Atmospheric levels of PBDEs show significant variation in time and space. Lillehammer stands out as a source area to the air around Lake Mjøsa for key compounds that are part of the technical mixture penta-BDEs. Unlike PCBs, the passive air samplers show that the general burden level of penta-BDE compounds over time is even higher in Lillehammer than in Oslo. The elevated levels appear to be limited to the Lillehammer region, which indicates that *in general* the emissions have limited potential for dispersal via air from Lillehammer to southern parts of Lake Mjøsa. The current data does not make it possible to say which source/s lead to a higher atmospheric burden in Lillehammer. It may be due to evaporation from highly-contaminated media (caused by past discharges) or there may be one or more primary sources in the Lillehammer region that have not been identified.

Based on the air measurements of $\Sigma PBDE_5$ (BDE-28, 47, 99, 100 and 153) in Kise, $\Sigma PBDE_5$ more than PCBs appears to be controlled by transport episodes with increased levels. An analysis of the origin of the air masses, and results assessed as a whole, indicates that the episodes of $\Sigma PBDE_5$ in Kise are due to both local atmospheric transport from the Lillehammer region and long-range transport (for which Britain seems to be a potential key contributor). It is interesting to note that the level of deca-BDE (BDE-209) in the air in Kise has the same level as of $\Sigma PBDE_5$, although BDE-209 is less volatile, and generally is believed to have a more limited potential for atmospheric long-range transport than other BDE compounds. The highest level of BDE-209 measured coincides with a long-range transported episode that also showed increased levels of $\Sigma PBDE_5$, where Britain appears to be a likely source region.

7.3 Mercury

The total mercury concentrations in precipitation in Kise are somewhat higher than in Birkenes. Due to a significantly higher amount of precipitation in Birkenes, the deposition in Kise is only half the level measured in Birkenes. Examinations of moss, combined with atmospheric emission data from Statistics Norway, indicate that the Lake Mjøsa region is not a significant source region of mercury emissions to the atmosphere. However, one cannot disregard the possibility that sources affecting the concentrations in air exist. A rough estimate of atmospheric deposition to Lake Mjøsa is 2 kg per year. An indicative level for sources from treatment plants is in the range 0.16–3 kg per year.

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STIKKORD Miljøgifter	Mjøsa	Modell	

REFERAT

Tilførsler av utvalgte miljøgifter til Mjøsa har blitt undersøkt og vurdert. Samlet vurdert tilsier resultatene at tilførselsmønsterert for PCB er sammensatt, og at det kan være betydelige kilder som ennå ikke er identifisert. Resultatene tyder imidlertid på at atmosfæren er en viktig kilde av PCB til Mjøsa, sammenlignet med en første beregning av tilførsler via elver og renseanlegg. Nivåene av PCB i luft rundt Mjøsa er forøvrig nokså typisk for bakgrunnsbelastningen i Norge. Forhøyede nivåer av PCB i luft tilskrives lokaltransport fra Osloregionen og langtransport fra andre land. Resultatene for kvikksølv indikerer også at de atmosfæriske tilførsler til Mjøsa primært skyldes langtransport fra områder utenfor regionen. Forhøyede nivåer av lavbromerte BDE-forbindelser i luft tilskrives utslipp i Lillehammerområdet og langtransport fra Storbritannia. Det generelle belastningsnivået av utvalgte BDE-forbindelser $\Sigma PBDE_5$ (BDE-28, 47, 99, 100 og 153) i luft ved Mjøsa er nå på nivå med BDE-209.

TITLE

An initial survey of the sources of contaminants to Lake Mjøsa.

ABSTRACT

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 results of this project furthermore suggest that the atmosphere is a key source of PCBs to lake Mjøsa, in comparison to preliminary estimates of the amounts discharged by rivers as well as sewage treatment plants. Elevated atmospheric levels of PCBs and PBDEs are in part attributed to long-range atmospheric transport of these compounds from other countries, whilst elevated levels of PBDE at Lillehammer suggest a local source to the atmosphere. The results finally indicate that the atmospheric deposition of mercury may primarily be explained by atmospheric long-range transport.

** Kategorier:* A *Åpen - kan bestilles fra NILU*
B *Begrenset distribusjon*
C *Kan ikke utleveres*