

Atmospheric deposition and lateral transport of mercury in Norwegian drainage basins:

A mercury budget for Norway



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REPORT

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<p>Summary</p> <p>Reduced atmospheric loading of mercury (Hg) can improve the environmental status of freshwaters and coastal ecosystems. Sparse data limit impact assessments of changes in atmospheric Hg on aquatic ecosystems. Here, a Hg budget is calculated for Norway by estimating fluxes of Hg in the environment. Atmospheric inputs (ca. 3.3 tonnes annually) are a factor 10 higher than riverine export to coastal areas, indicating considerable retention in soils and lakes. Environmental loading of Hg to surface waters was separated into headwater (=catchment) export of Hg and atmospheric Hg loadings, which were of similar size (0.2 to 0.3 tonnes), where atmospheric loadings were considered of higher certainty than headwater fluxes. Robust methods for estimations of headwater Hg export at the regional scale are lacking. Hg budgets for small headwater catchments are more reliable and valuable to assess the fate of atmospheric Hg, and the role of aquatic Hg retention. Continued monitoring of Hg in deposition, catchments, and rivers are needed to reduce uncertainties in environmental assessments, in addition to international collaboration.</p>

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Preface

Mercury is a natural element present all over the world, which can be mobilised through both anthropogenic activities and natural processes. Assessments of the fate of mercury in the environment are often limited by lack of data and our understanding of the mercury cycling processes. These knowledge gaps restrict environmental impact evaluation of applied mercury emission reduction measures under different environmental agreements such as the *Minamata Convention on Mercury* and the *Convention on Long-Range Transboundary Air Pollution*. Here, we address some of the knowledge gaps by estimating the fate and transport of mercury in the Norwegian environment, and by calculating an overall mercury budget. Our aim is to assess data quality and uncertainty, and to evaluate how assessments of Hg loading to aquatic ecosystems can be improved.

The work is funded by the Norwegian Environment Agency (NEA), and has received support from several other initiatives, including: The Norwegian Institute for Water Research's (NIVA's) Strategic Institute Initiative (SIS) "*Land-Ocean Interactions*" (project number #180033); and the International Cooperative Programme on Integrated Monitoring (www.syke.fi/nature/icpim). Additionally, we would like to thank the project Climer (#243644), funded by the Research Council of Norway, and NIVA's core funding for monitoring of mercury in the Norwegian environment.

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Oslo, 07.12.2018



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Summary

Mercury (Hg) is a natural element, present all over the world at trace concentrations. The element has a long lifetime in the atmosphere, and nearly two centuries of elevated Hg deposition from anthropogenic activities have led to considerable stores of Hg in catchment soils and high concentrations in aquatic food webs, even in remote locations. In Norway, concern relates to elevated concentrations of Hg in freshwater fish, many places exceeding maximum limits for protection of human health. To allow for effectiveness evaluations of Hg emission reduction measures on Hg in aquatic ecosystems under different environmental agreements such as the *Minamata Convention on Mercury* and the *Convention on Long-Range Transboundary Air Pollution*, assessments of the fate of Hg in the environment are necessary. However, these evaluations are often limited by lack of data and our understanding of the Hg cycling processes. Here, we estimate a Hg budget for Norway, by quantifying key fluxes to Norway, including deposition, and lateral fluxes from soils (headwaters) to surface waters and by rivers to the coast. Our aim is to contribute to a qualitative assessment of data quality and uncertainty, and to evaluate how assessments of Hg loading to aquatic ecosystems can be improved.

Based on modelled deposition under the European Monitoring and Evaluation Programme (EMEP) from 2016, we estimate a total annual deposition of 3077 kg ($9.5 \mu\text{g m}^{-2}$) of Hg to Norwegian land. Of this, 2488 and 589 kg are deposited through wet and dry processes. An independent estimate for Hg deposition in forests based on national estimates of litterfall and throughfall (forest monitoring data), combined with concentration data, amounted to only 40% ($5.6 \mu\text{g m}^{-2}$) of EMEP estimated total Hg deposition in forests ($13.5 \mu\text{g m}^{-2}$). For the most densely forested part of Norway (i.e. the Skagerrak drainage basin), the EMEP ($9 \mu\text{g m}^{-2}$) and forest monitoring ($10 \mu\text{g m}^{-2}$) estimates were more similar. The upscaling of data from forest monitoring to the whole of Norway is uncertain for high-elevation and high-latitude areas with open, low productivity forest that gradually transitions to shrublands and tundra because litterfall and throughfall are difficult to estimate. We conclude that the estimates from EMEP and the upscaling of forest monitoring data are in reasonable agreement with each other, but that both estimates would benefit from validation with more empirical data.

We used data from the national river monitoring programme to estimate riverine Hg export to coastal areas. Two methods of calculation resulted in similar estimates of riverine Hg export, i.e. 281 ± 84 (derived from riverine carbon export) and 331 ± 87 kg (based on riverine Hg transport, means \pm one standard deviation of annual data). We judge the coastal inputs of Hg to be relatively reliable compared with other flux estimates presented in this report, because both components (i.e. runoff and concentrations) have an empirical basis. Ratios of Hg-to-TOC (total organic carbon) showed limited variation between rivers and lakes, suggesting that Hg and organic carbon cycling are closely linked. Given an analytical method with a lower detection limit for Hg than currently applied, inter-annual and long-term change in riverine Hg export can be quantified with the current river monitoring programme. Only circa 10% of the estimated atmospheric inputs were exported as riverine Hg input to the sea. This is likely a reflection of retention of Hg in catchment soils, along the aquatic continuum from soils to coast, and from re-emission of deposited Hg to the atmosphere. Soil retention and re-emission of Hg are poorly understood and difficult to quantify.

The lack of reliable estimates of headwater Hg inputs limit our ability to quantify Hg retention along the aquatic continuum, before Hg export to the coast. With data on organic carbon generally being more available than for Hg, ratios of Hg-to-carbon can be used to estimate Hg fluxes based on the organic carbon data. We created a simple model linking headwater organic carbon export to runoff, forest cover, and peatland cover (for a subset of Norwegian catchments) and up-scaled the findings using the Regine dataset for all headwater catchments in Norway. The carbon fluxes were multiplied with a Hg-to-carbon ratio from lake and river monitoring, resulting in a mass flux of 295 kg Hg from headwaters. The estimated headwater export is similar to the riverine export of Hg (281-331 kg), suggesting that retention of Hg along the aquatic continuum is of similar size as the direct atmospheric inputs to surface waters (251 kg). However, as the estimate of the headwater TOC export is uncertain, caution should be applied in the interpretation of this. Still, the similarity of the catchment loading and surface waters deposition estimates could imply that reductions in wet deposition of Hg would have positive implications for Hg accumulation in the aquatic foodweb. A more process-based headwater TOC model would likely result in better estimates of headwater TOC export, but is currently not available. Thus, environmental loading of Hg to surface waters from catchments and atmosphere is very difficult to quantify at the regional scale, which limit assessments of how changes in anthropogenic emissions affect aquatic ecosystems. For catchment case studies (headwaters), such processes are more easily studied with a well-designed monitoring programme.

All data were synthesized in a Hg budget for Norway, expressed in mass and in mass per unit area. The dominating origin of Hg in the Norwegian environment is air pollution rather than industrial point sources. Norway receives a factor 10 more Hg by air pollution than is exported to coastal waters. Large knowledge gaps relate to re-emission of deposited Hg, catchment inputs of Hg to headwaters and retention along the aquatic continuum. Whether (or at what rate) reduced emissions of Hg to the atmosphere will result in lower loading of Hg to surface waters (and therefore reduced exposure of aquatic ecosystems to Hg) is difficult to assess, especially given the large stores of legacy Hg present in soils and lake sediments and the complexity of Hg mobilization and transport mechanisms.

Given the limited understanding of Hg cycling and availability of monitoring data for deposition, catchment export, and retention of Hg, we recommend continuation of catchment and river monitoring of Hg, and stimulation of international collaboration on Hg monitoring and modelling. The quantification of key elements in the Hg budget for Norway is useful for highlighting available data sources and data gaps, in addition to the highlighted limited ability to compare independent estimates of Hg fluxes obtained from various sources and with different approaches. We recommend continued collection of monitoring data as the most reliable way to ground-truth modelled estimates of Hg supply, fate, and effects.

Sammendrag

Kvikksølv (Hg) er et naturlig grunnstoff som finnes i lave konsentrasjoner i naturen. Grunnstoffet har lang levetid i atmosfæren, og nesten to århundrer med høye antropogene Hg-utslipp har ført til store lager i jord samt høye konsentrasjoner i akvatiske næringskjeder, selv i områder langt unna punktutslipp. En stor bekymring i Norge er høye konsentrasjoner av Hg i ferskvannsfisk. Mange steder er konsentrasjonene høyere enn gjeldende grenseverdier. For å kunne vurdere effekten fra ulike Hg-reduserende tiltak under internasjonale avtaler slik som *Minamatakonvensjonen* og *Konvensjonen om langtransportert luftforurensing*, er evalueringer av skjebnen til Hg i miljøet nødvendig. Slike evalueringer er imidlertid ofte begrenset av mangel på data samt forståelsen av Hg-prosesser i miljøet. I denne rapporten har vi estimert et Hg-budsjett for Norge ved å kvantifisere viktige transportprosesser som inkluderer atmosfærisk deponisjon, transport fra nedbørfelt til innsjøer, og transport via elvene til kystområder. Målet har vært å bidra til en kvalitativ vurdering av datakvalitet og usikkerhet, og hvordan vurderinger av Hg-tilførsler til akvatiske økosystemer kan forbedres.

Basert på modellerte deponisjonstall fra European Monitoring and Evaluation Programme (EMEP) for 2016, har vi estimert en total årlig avsetning på 3077 kg ($9.5 \mu\text{g m}^{-2}$) Hg til Norge. Av dette er 2488 og 589 kg deponert som henholdsvis våt- og tørrdeponisjon. Til sammenlikning ga et uavhengig estimat for atmosfærisk Hg-avsetning til norske skogsområder, basert på nasjonale skogovervåkingsdata for strøfall og nedbør gjennom trekronene, en total Hg deponisjon som kun utgjorde omlag 40 % ($5.6 \mu\text{g m}^{-2}$) av den EMEP-estimerte avsetningen til skogsområder ($13.5 \mu\text{g m}^{-2}$). Imidlertid samsvarte de to metodene bedre ved kun å se på områdene i Norge med tettest skog (EMEP: $9 \mu\text{g m}^{-2}$ og skogovervåkingen: $10 \mu\text{g m}^{-2}$, områdene som drenerer til havområdet Skagerrak). En oppskalering av data fra skogovervåkingen til hele Norge er forbundet med betydelig usikkerhet for nordlige og høyereliggende områder med åpen, lavproduktiv skog som gradvis endres til krattskog og tundra. Dette fordi strøfall og nedbør gjennom trekronene er vanskelig å estimere for disse områdene. Vi konkluderer med at estimatene fra EMEP og skogovervåkingen stemmer brukbart overens, men at begge datakildene med fordel kunne vært validert med empiriske data.

For å estimere elvetransport av Hg til norske kystområder ble data fra det nasjonale Elveovervåkingsprogrammet benyttet (tidligere Elvetilførselsprogrammet). To ulike metoder resulterte i relativt like estimater: 281 ± 84 kg (basert på elvetransport av karbon) og 331 ± 87 kg (basert på elvetransport av Hg, gjennomsnitt \pm ett standardavvik for årlige data). Vi vurderer disse nivåene av Hg-transport med elver som robuste sammenlignet med andre fluksestimater fra denne rapporten, ettersom begge komponentene i analysen (avrenning og konsentrasjoner) er basert på empiriske data. Forholdet mellom konsentrasjoner av Hg og totalt organisk karbon (TOC) varierte lite mellom elver og innsjøer, noe som indikerer at Hg-syklusen i miljøet er tett knyttet til kretsløpet for organisk karbon. Det er bemerkelsesverdig at bare ca. 10 % av den estimerte atmosfæriske avsetningen av Hg til Norge ble eksportert til kystområdene med elvene. Dette kan sannsynligvis forklares ved stor grad av Hg-retensjon i jord, fra nedbørfelt til kystområder, og ved re-emittering av allerede avsatt Hg tilbake til atmosfæren. Retensjon i jord og re-emittering er to prosesser som er mangelfullt forklart i litteraturen og vanskelig å kvantifisere. For å kunne kvantifisere årlige- og langtidsendringer av Hg-eksport fra

norske elver bør en kjemisk analysemetode med lavere deteksjonsgrense enn den som i dag benyttes bli inkludert i det nasjonale Elveovervåkingsprogrammet.

Mangelen på pålitelige estimater av hvor mye Hg som transporteres fra små primære vann øverst i vassdraget (her kalt utspringssjøer), gjør det vanskelig å kvantifisere retensjonen av Hg mellom atmosfærisk avsetning og eksport til kystområder. Data for organisk karbon er ofte mer tilgjengelig enn data for Hg, og vi har derfor benyttet forholdet mellom konsentrasjoner av Hg og TOC til å estimere flukser av Hg fra slike utspringssjøer. En enkel lineær modell ble utviklet, der vi koblet eksport av organisk karbon fra utspringssjøer til avrenning og andelen skogs- og myrområder for et utvalg norske nedbørfelt. Resultatene ble oppskalert ved å benytte Regine-databasen for alle utspringssjøer i Norge. Basert på dette ble karbonflukser multiplisert med et forholdstall for Hg-til-karbon fra elve- og innsjøovervåking. Resultatet ble at ca. 295 kg Hg eksporteres fra norske utspringssjøer. Denne mengden Hg er veldig lik mengden Hg som transporteres med elvene til norske havområder (281-331 kg), hvilket betyr at retensjonen av Hg i vassdragene (altså i elver og innsjøer) er av omtrent samme størrelse som den direkte atmosfæriske avsetningen av Hg til innsjøer (251 kg). Estimaterne av karbonfluksene fra utspringssjøene er imidlertid usikre og tolkningene av dataene bør være deretter. Allikevel kan den lille forskjellen i estimater av Hg-eksport fra utspringssjøer og atmosfærisk avsetning til innsjøer antyde at redusert våtdeposisjon av Hg vil ha positive effekter på akkumulering av Hg i akvatiske næringskjeder. En tydeligere prosessbasert modell for karbon i disse innsjøene ville sannsynligvis resultere i bedre estimater for karboneksporten, men det er ikke tilgjengelig nå. Som en følge av dette er det vanskelig å kvantifisere belastningen av Hg til innsjøer fra henholdsvis nedbørfelt og atmosfære på regional skala, noe som igjen begrenser vurderingen av hvordan antropogene utslipp av Hg påvirker akvatiske økosystemer. Imidlertid kan studier av slike prosesser på nedbørfeltnivå være enklere å studere ved for eksempel et godt designet overvåkingsprogram.

All data ble sammenstilt til et Hg-budsjett for Norge. I Norge er langtransportert luftforurensing en mye større kilde til Hg i miljøet enn industrielle punktkilder, og eksporten med elver til kystområdene utgjør bare omtrent 10 % av atmosfærisk avsetning. Imidlertid er det store kunnskapshull relatert til re-emittering av allerede avsatt Hg, tilførsler av Hg til innsjøer fra nedbørfelt, og retensjon av Hg fra nedbørfelt til kystområdene. Hvorvidt reduserte utslipp av Hg til atmosfæren vil resultere i lavere belastning av Hg til innsjøer (og derfor redusert Hg-eksponering for akvatiske økosystemer) er vanskelig å vurdere, spesielt på grunn av de betydelige lagrene av Hg i jord og innsjøsedimenter, og på grunn av de kompliserte mobiliserings- og transportmekanismene for Hg i miljøet.

Gitt den begrensede forståelsen av Hg-syklusen og tilgjengeligheten av Hg-data for deposisjon, nedbørfelteksport og retensjon av Hg, anbefaler vi en fortsettelse av overvåking av Hg i norske nedbørfelt og elver, og samtidig en økt stimulering av internasjonalt samarbeid på overvåking og modellering av Hg i miljøet. Kvantifiseringen av et Hg-budsjett for Norge er nyttig for å kunne belyse tilgjengelige datakilder og kunnskapshull, i tillegg til begrensningene som ligger i å sammenligne uavhengige estimater av Hg-flukser basert på ulike kilder og tilnærminger. Vår anbefaling er at fortsatt innsamling av overvåkingsdata er den mest pålitelige kilden til empiriske modellestimater for tilførsel, skjebne og effekter av Hg.

Tittel: Atmosfærisk deposisjon og transport av kvikksølv i norske nedbørfelt – Et kvikksølvbudsjett for Norge

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1 Introduction

1.1 Background and status of knowledge

Mercury (Hg) is a natural element, present all over the world at trace concentrations. The element has a long lifetime in the atmosphere (approximately 1 year as gaseous elemental Hg, Hg⁰) (Slemr et al., 2003), and nearly two centuries of elevated Hg deposition from anthropogenic activities (Streets et al., 2011) have led to considerable stores of Hg in catchment soils, even in remote locations (Fitzgerald et al., 1998).

Hg in its organic form, as methyl-Hg (MeHg), is biomagnified in aquatic food webs and has proven to have harmful effects on aquatic organisms (Wolfe et al., 1998) and their consumers (Scheulhammer et al., 2007), including humans (Driscoll et al., 2013; Zahir et al., 2005). In 1956, the Minamata Bay accident initiated monitoring and research on Hg (Kurland et al., 1960). Since the occurrence of the accident, researchers have struggled to predict impacts of changed Hg emissions on exposure, accumulation, and biomagnification of Hg in food webs, mainly because of the complex biogeochemical cycling of the element (Sonke et al., 2013). Thousands of freshwater lakes worldwide have fish Hg concentrations exceeding limits advised for human consumption (0.3 – 1.0 ppm Hg wet weight (w.w.)) (UNEP, 2002), including many lakes in Norway (Braaten et al., 2017). Freshwater fish are considered to be critical receptors of long-range transboundary air pollution of Hg (CLRTAP, 2016).

The *Minamata Convention on Mercury* (hereafter *Minamata Convention*) aims to protect human health and the environment from adverse effects of Hg at a global scale (UNEP, 2014). The agreement requires the parties to evaluate its effectiveness, based on “*comparable monitoring data on the presence and movement of Hg and Hg compounds in the environment*” under Article 22. The Minamata Convention was signed by 93 countries in 2013. In early 2017, the 50-ratification milestone required for the convention to enter into force was reached, and later that same year the agreement became legally binding for all its parties (UNEP, 2014).

For evaluation of applied Hg emission reduction measures under different environmental agreements such as the Minamata Convention and the *Convention on Long-Range Transboundary Air Pollution* (CLRTAP), assessments of effects on the environment is necessary. However, knowledge of processes occurring between deposition and aquatic transport is limited, partly because of limited availability of relevant data. This poses challenges for the evaluation of how changes in Hg emissions affect Hg exposure of organisms in aquatic environments.

1.1.1 Mercury speciation

In the environment, Hg can be found as several different species, of varying toxicity, mobility, and bioaccumulation potential. In short, Hg can exist in its elemental, inorganic, or organic form (Leermakers et al., 2005). Hg⁰ is the most stable form in the atmosphere and can be transported over long distances (Schroeder et al., 1993). Inorganic Hg (Hg⁺ and Hg²⁺) constitutes the major Hg species in terrestrial and aquatic environments, and the in its ionic form Hg has a strong affinity for various inorganic and organic ligands (Leermakers et al., 2005). Organic Hg (CH₃Hg⁺ and (CH₃)₂Hg) is commonly

referred to as methyl-Hg (MeHg) (Leermakers et al., 2005), and this constitutes the toxic and bioaccumulative form in aquatic ecosystems (Clarkson and Magos, 2006).

1.1.2 Mercury in the atmosphere and deposition

In the atmosphere, elemental and reactive gaseous Hg (RGM), in addition to Hg bound to particles (Hg-P) are the three major species (Schroeder and Munthe, 1998). Atmospheric concentrations of Hg in the Northern hemisphere are typically in the range of 1.5 - 1.7 ng m⁻³, and dominated by 97% gaseous elemental Hg (Slemr et al., 2003). Intercontinental transport from Hg emission sources outside Europe is the main factor responsible for the current air concentrations in Europe. Because of this, atmospheric concentrations of Hg are relatively consistent within Europe (1.4 – 1.6 ng m⁻³) (EMEP, 2016). In the European region, atmospheric concentrations have decreased with 23% from 1990 to 2014 (Streets et al., 2011), and most of the decrease occurred before year 2000 (EMEP, 2016).

The flux of Hg from the atmosphere to the earth's surface occurs through wet or dry deposition. Wet deposition is washout of Hg by precipitation (i.e. rain, snow, fog, ice), while dry deposition occurs in the absence of precipitation (Lindberg et al., 2007). Both wet and dry deposition are influenced by emission sources, regional atmospheric chemistry and (micro-) meteorological conditions (Lindberg and Stratton, 1998). In Europe, average total deposition was around 12 µg m⁻² year⁻¹ in 1990 and just above 10 µg m⁻² year⁻¹ in 2014 (EMEP, 2016). While the analytical determination of Hg in wet deposition is well established, measurements of Hg concentrations in air and dry deposition are limited, and atmospheric deposition is often estimated by numerical models (e.g. regional scale modelling of the European region done by *European Monitoring and Evaluation Programme*) (EMEP, 2016)). These models depend largely on meteorology, including temperature, wind speed, and surface wetness and geomorphology, and actual measurements remain a large knowledge gap (Lindberg et al., 2007).

For dry deposition of Hg⁰, RGM, and Hg-P, vegetation plays an important role (Grigal, 2003). Because of the large surface area of receptor sites on plant leaves, dry deposition is typically more important in forested areas than in open areas (Graydon et al., 2012). Hg⁰ is typically absorbed in vegetation and deposited via litterfall, while RGM and Hg-P adsorb to surfaces and can also be washed to the ground as throughfall (Lindberg et al., 1994). Total Hg deposition in forests can be estimated as the sum of litterfall and throughfall. Dry fluxes of Hg from the atmosphere can be approximated as the sum of litterfall and net throughfall (i.e. throughfall Hg flux minus precipitation Hg flux) (Munthe et al., 1995).

1.1.3 Mercury catchment cycling and the role of organic matter

Deposited Hg is associated with low mobility and accumulates in catchment soils. Nearly two centuries of elevated deposition have led to considerable stores in soil, even at remote locations (Fitzgerald et al., 1998). Inorganic Hg can be methylated to the neurotoxic MeHg (Bloom, 1992; Gilmour et al., 1992), potentially harming organisms and humans (Scheulhammer et al., 2007). Both inorganic and organic Hg species can form association with various forms of organic matter (OM) (Driscoll et al., 1995; Mierle and Ingram, 1991), which is explained by Hg having a high affinity for sulphur (S) sites on the OM (i.e. thiol groups) (Ravichandran, 2004; Skyllberg et al., 2006). The mobility of Hg can increase by forming

association with the dissolved fraction of the OM (DOM < 0.45 μ m), which can be considered as the main transport vector for Hg (and MeHg) from soils to streams and lakes (Grigal, 2002).

The strong relationship between Hg and OM in lake and stream waters has been documented spatially over the boreal forest zone, illustrated in several Scandinavian (Braaten et al., 2014a; Eklof et al., 2012; Meili et al., 1991; Skyllberg et al., 2003) and North American (Benoit et al., 2003; Driscoll et al., 1995; Shanley et al., 2008) studies. Interestingly, fluxes of Hg in lake outlets are usually modest relative to the catchment storage of Hg (Grigal, 2002; Grigal, 2003). This suggests that leaching of Hg from soils is likely to be an important source of Hg in surface waters (lakes and streams) for decades or centuries to come (Larsen et al., 2008).

1.1.4 River transport of mercury

Riverine Hg inputs are a significant source of Hg to the North Atlantic and the Atlantic Ocean (Driscoll et al., 2013; Fisher et al., 2012), and constitutes around 10% (380 Mg year⁻¹) of global atmospheric deposition to the ocean (4000 Mg year⁻¹) (Driscoll et al., 2013). Limited information is available regarding spatial and temporal variation in riverine Hg loadings to the ocean and how coastal ecosystems are impacted by these inputs. Recent investigations suggest that the riverine inputs have declined in the last few decades (Sunderland et al., 2012).

A challenge with analysing Hg in river systems is the limited time series available for measurements of low-level Hg concentrations. However, from the few studies available, some predictors for Hg concentration have been shown. For example, strong correlations between particulate Hg and particulate fractions of OM (Regnell et al., 2009) and total suspended solids (TSM) (Balogh et al., 2008) have been documented. Also, run-off concentrations of Hg have been shown to be influenced by hydrology: in a study of North American streams, discharge was found to be the physical variable explaining most of the Hg variation (Brigham et al., 2009). In fact, Brigham et al. (2009) suggest that discharge, dissolved organic carbon (DOC), particles, and wetland abundance can be used for predicting concentrations of Hg in rivers.

Although a yearly flux from terrestrial Norway to the ocean is calculated as part of the Norwegian obligations under the Oslo-Paris Convention (OSPAR), an estimation of temporal trends has not been undertaken due to many samples being reported below the detection limit (Skarbøvik et al., 2017).

1.2 Main report goals and objectives

The fate of anthropogenic emissions of Hg in the environment, and the impact of environmental policy to reduce such emissions, is difficult to assess at national scales. Especially for surface waters it is important to understand how reductions of Hg to the environment can impact Hg exposure of organisms in aquatic environments. Fish constitute an important food source for humans and wildlife, and is a main route of exposure. Loading of Hg to aquatic ecosystems occurs through atmospheric deposition, catchment export and mobilization from sediments. Currently, a qualitative assessment of data quality and uncertainty to quantify such loadings is lacking, which limits our ability to evaluate impacts of changes in Hg loading to aquatic ecosystems.

Measurements of Hg from monitoring stations require upscaling to the appropriate level for a national evaluation. However, such upscaling exercises of Hg in the environment is often limited by *i)* lack of data; *ii)* our understanding of the Hg cycling processes; and *iii)* methodological uncertainties (i.e. lack of validation by comparison with direct observations from monitoring programs). As an example, modelled wet and dry deposition of Hg from EMEP may not be an accurate estimate of total atmospheric inputs of Hg to Norway, because of uncertainties in estimation of interception of Hg in forests, where dry Hg deposition is likely to be a significant deposition pathway (Graydon et al., 2008). Therefore, it is important to increase our knowledge of the relative contribution of Hg to waters from deposition and catchments, the transport of Hg through the catchments, and the discharge of Hg out of the catchments, to allow for an evaluation of reduced Hg emissions effects on the environment.

Here, we aim to address some of these knowledge gaps by calculating and estimating fluxes of Hg in the Norwegian environment, with a focus on processes controlling transport from headwater catchments to the rivers discharging in the sea. Because of the close connections between Hg and carbon cycling, and the much larger data availability for carbon in terrestrial and aquatic ecosystems, we will aim to link Hg to carbon to constrain uncertainties in the estimated fluxes of Hg. We aim to establish a national Hg budget for Norway. The following main environmental Hg transport processes are evaluated (**Figure 1**): (1) atmospheric Hg deposition; (2) Hg from catchments to headwaters; (3) retention and processing of Hg in the aquatic continuum; and (4) Hg river export from Norway to the sea.

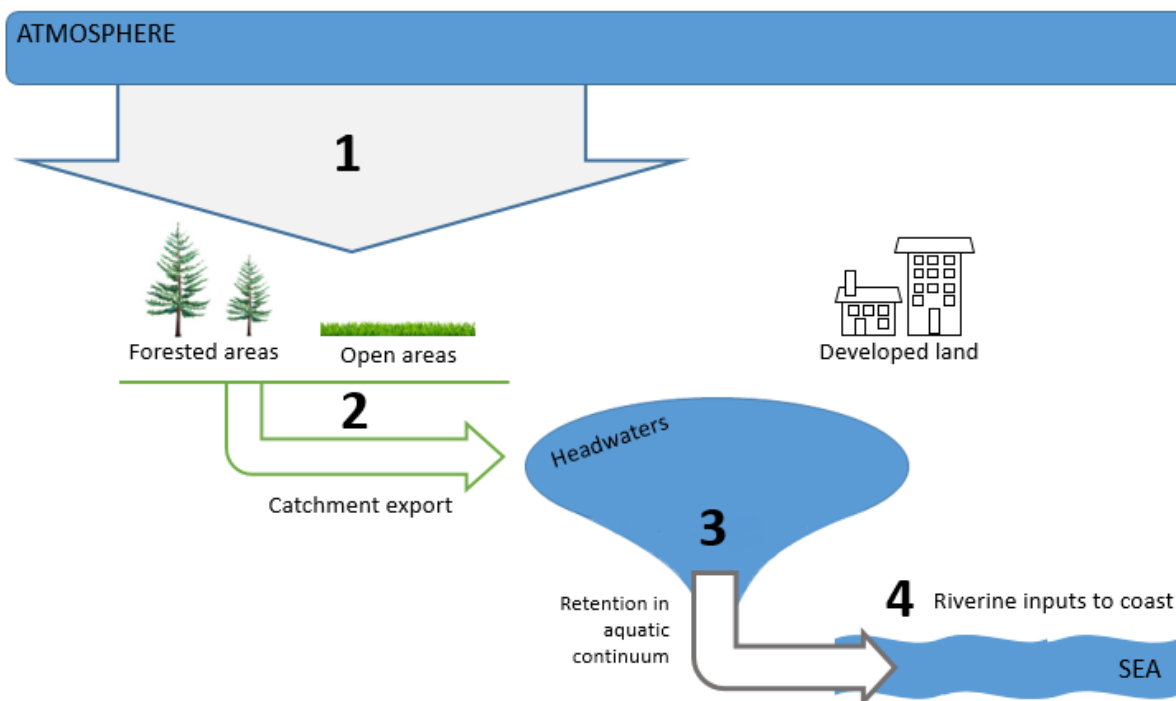


Figure 1 A sketch of the processes discussed in this report, including (1) atmospheric Hg deposition; (2) catchment export of Hg; (3) aquatic processing of Hg; and (4) riverine inputs to the coast.

The scope of the work presented includes estimates of environmental transport processes for Hg under natural conditions, i.e. anthropogenic releases such as Hg in wastewater are not accounted

for. Additionally, effects from natural processes potentially affecting transport flux of Hg to the sea, e.g. coastal erosion, are not included.

2 Materials and methods

2.1 Geographical extent and limitations

The Norwegian mainland drains to four regions (**Figure 2**), including (from south to north) Skagerrak (from the Swedish border to the southernmost point of Norway at 57° 44'N), the North Sea (from 57° 44'N to 62° N), the Norwegian Sea (from 62° N to 68° 15'N), and the Barents Sea (from 68° 15'N to the Russian border). These four regions are used when Norway are reporting riverine inputs and discharges to coastal waters as part of the obligations under the OSPAR Convention (i.e. the European mechanism to protect the marine environment of the North-East Atlantic). Additionally, the division into four regions was used in 2015 by De Wit et al. to establish a carbon budget for Norway (de Wit et al., 2015). We have chosen to follow this grouping in the present work as it will enable an upscaling of findings from local and regional studies within each region to a larger area, i.e. to do a source apportionment for Hg within each region.

2.2 Estimation of atmospheric deposition of Hg

EMEP is a programme under the CLRTAP for international cooperation to solve transboundary air pollution problems. EMEP includes five centres, where the Meteorological Synthesising Centre – East (MSC-E) is responsible for heavy metals (including Hg) and POPs (MSC-E, 2018).

In modelling of Hg deposition, EMEP uses meteorology and official country-specific emissions of Hg. Observational data of atmospheric Hg concentrations and deposition are used to validate the modelling results, but the spatial and temporal resolution of such data are not sufficient to serve as a model input. The atmospheric removal processes for Hg includes both dry deposition and wet scavenging (i.e. removal by precipitation) (MSC-E, 2018). For Hg, the EMEP data includes Hg species in the solid, gaseous, and aqueous phases, including particulate Hg (Hg-P); gaseous Hg (Hg_{gas}, including Hg⁰ and Hg²⁺ on gaseous form); and dissolved Hg (Hg_{diss}, including dissolved Hg⁰, Hg-sulphate HgSO₄, and Hg-chloride HgCl₂). In this work, we have focused on separating wet and dry Hg deposition to different land cover, i.e. forested areas, waters, open areas, and developed land) for the part of the EMEP-grid (50 x 50 km spatial resolution) that covers the Norwegian mainland for the year 2016.

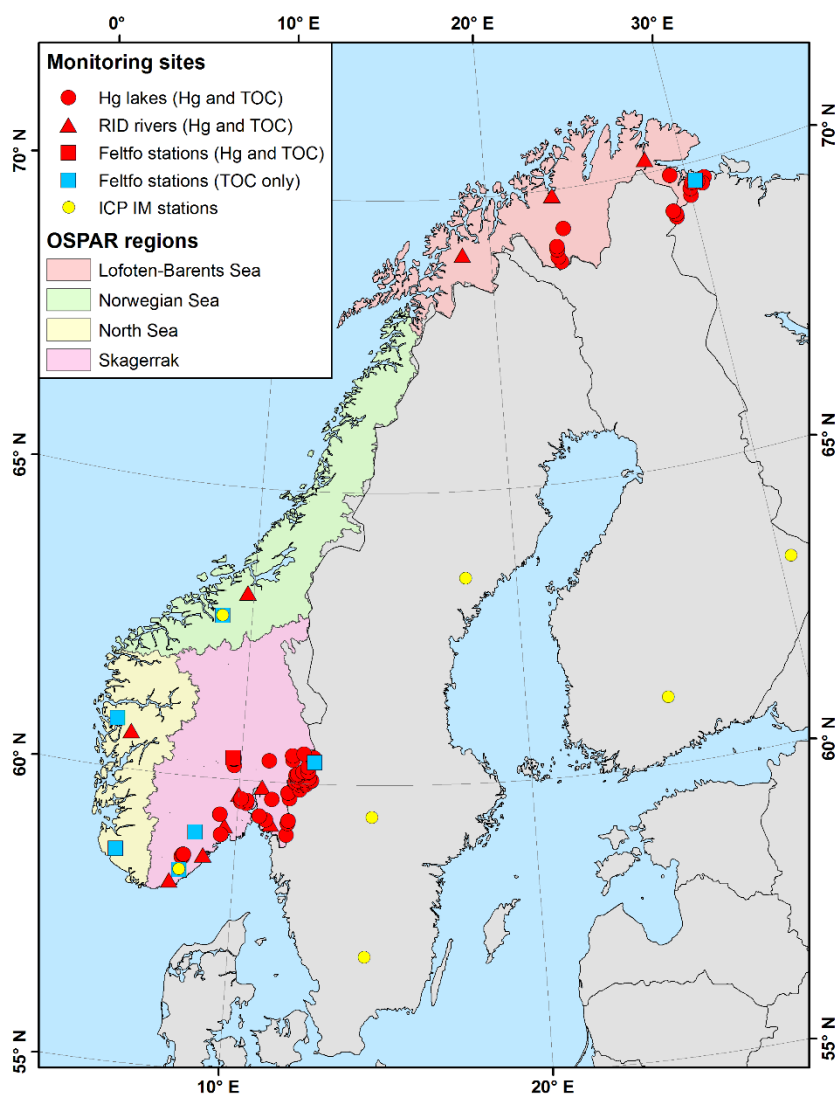


Figure 1 Norway is divided into four main drainage areas, which from south to north drain into the Skagerrak, the North Sea, the Norwegian Sea, and the Barents Sea. Minor parts of Norway drain to Sweden. On the map is also indicated the location of the sampling stations utilised for Hg and TOC water chemistry analysis, including stations for Hg and TOC in lakes (red circles, n=70); river monitoring (“RID”) stations (red triangles, n=11); and headwater catchment stations (“Feltfo stations”) where some include Hg and TOC data (red square, n=1, Langtjern) and some only TOC data (blue squares, n=7). Stations from ICP Integrated Monitoring, providing Hg deposition and runoff data, are indicated as yellow circles.

2.3 Estimation of forest Hg deposition from litterfall and throughfall

Total Hg deposition to forests can be approximated from measurements of litterfall and throughfall. This was done in two steps: 1) concentrations of Hg in litterfall and throughfall at four extensively studied forested case sites in Norway and Sweden (ICP Waters Integrated Monitoring, IM) was assessed, and 2) the Hg litterfall and throughfall concentrations were extrapolated to national scale based on available forest descriptors (National forest inventory), and throughfall and precipitation data.

1) Case study of Hg in litterfall and throughfall

The case studies compiled three sites in Sweden (Aneboda, Kindla, and Gammtratten) and one in Norway (Langtjern), which have previously been established through ICP IM (Kleemola and Forsius, 2018). The study sites represent both pine (Langtjern) and spruce forests (Aneboda, Kindla, Gammtratten, and Langtjern), and are located along a north to south gradient (**Figure 2**). This allows for a consideration of forest types and latitudinal geographical location as predictors for variation in Hg concentration.

2) Extrapolating Hg litterfall and throughfall deposition to national scale

To allow for an upscaling of Hg litterfall and throughfall from the case studies to the whole of Norway, national estimates of total litterfall and throughfall was needed. An estimate of total litterfall in Norwegian forests was available from (de Wit et al., 2015) in which a national carbon budget for Norway was established. In short, litterfall was estimated based on stem volume data (Norwegian Forest Inventory) which was converted to needles and leaves via biomass. Measurements from a total of eight Norwegian sites (i.e. “*Feltfo*”-stations) were the basis for the calculations, which covered all four drainage basins of Norway (described under 2.1 **Figure 2**). For more information on litterfall calculations, see (de Wit et al., 2015). An estimate of national throughfall data was not available. To circumvent this, the relationship between throughfall and precipitation was utilised. Throughfall depends primarily on two factors, i.e. canopy density and precipitation intensity. Dense forests intercept more precipitation than open forests, which makes throughfall a more important process for Hg deposition in dense forests compared to open forests. Higher precipitation is likely to result in a lower fraction of precipitation that is intercepted, and evaporated. In high density forests in relatively dry regions, throughfall fluxes of Hg will be a relatively more important pathway for Hg loading in comparison to wet deposition, than in open forests in wet regions.

Based on averages of long-term measurements of throughfall and precipitation at the same eight forested sites that were the foundation for the litterfall estimate (“*Feltfo*”-stations), throughfall-to-precipitation ratios were calculated. The ratios are presented in **Table 1** and were calculated using both mean and median precipitation and throughfall, which resulted in very similar values ranging from 0.71 to 0.95. For the extrapolation of Hg litterfall and throughfall to a national scale, different ratios were used for the various sub-regions of the four drainage basins of Norway to consider factors such as precipitation intensity, high altitude, and geographical location. For example, the mean throughfall to precipitation ratio for the two wettest Norwegian sites (0.87) were used for the four wettest regions in the south, and the mean of the ratios for the four Swedish and Finnish sites (0.75) was used for the remaining sites in the south. For the Barents Sea and the high-altitude region (region 8) we use the throughfall ratio for the site with the lowest forest density, NO03 (0.95), which is the site with the lowest forest density. The annual mean litterfall and throughfall Hg fluxes were expressed in $\mu\text{g m}^{-2}$ forest. Total forest Hg deposition was calculated as the sum of throughfall and litter fluxes.

Table 1 Mean and median precipitation (P, mm), throughfall (TF, mm), and TF-to-P ratio for 7 ICP IM monitoring sites in Finland (FI01, Valkea-Kotinen; FI03, Hietajarvi), Norway (NO01, Birkenes; NO02, Kårvatn, NO03, Langtjern) and Sweden (SE14, Aneboda; SE15, Kindla).

Station code (latitude)	Years of data	Precipitation (P) (mm)		Throughfall (TF) (mm)		TF-to-P ratio	
		median	mean	median	mean	median	mean
FI01 (61.14)	15	618	628	430	444	0.71	0.71
FI03 (63.09)	15	613	631	566	579	0.91	0.92
NO01 (58.23)	17	1701	1719	1591	1496	0.87	0.87
NO02 (62.47)	11	1468	1529	1299	1291	0.86	0.85
NO03 (60.22)	4	848	940	810	911	0.95	0.96
SE14 (57.07)	17	859	826	616	598	0.73	0.75
SE15 (59.45)	17	873	914	586	598	0.66	0.66

2.4 Hg in Scandinavian lakes and streams

2.4.1 Lakes

Surface waters of pristine lakes (i.e. no local contamination source) in Norway typically shows concentrations in the pg to ng L⁻¹ range (Braaten et al., 2014a; de Wit et al., 2014). Studies of Hg species in surface waters of natural catchments in Scandinavia in general, and in Norway are rare. The largest gathering of data is a study of 51 lakes from southern and northern Norway from 2014 (Braaten et al., 2014a). In these natural pristine lake systems, OM is the main transport vector for Hg from catchment soils to surface waters (Grigal, 2002), and concentrations of total OC (TOC) and DOC typically correlate strongly with concentrations of Hg in lake surface water (Eklof et al., 2012; Meili et al., 1991; Skyllberg et al., 2003). Here, we have gathered data from available literature and combined it with previously unpublished data from NIVA to assemble a lake water database of Hg and TOC concentrations. In the database, the two variables are correlated for different Norwegian regions, and used to estimate Hg concentrations throughout Norway based on available TOC measurements.

2.4.2 Streams in Norway, Sweden, and Finland

Stream water data were obtained from monitoring programs in Norway, Finland, and Sweden. In Norway, eight headwater catchments (i.e. Birkenes (NO01), Storgama, Langtjern (inlet and outlet, NO03), Kårvatn (NO02), Svartetjern, Øygardsbekken, Dalelva) have been monitored since the 1970s and 1980s (Garmo et al., 2015), and in which TOC measurements started in the middle of the 1980s. Svartetjern is no longer an active station. Only in one Norwegian catchment, Langtjern, Hg data are available. The data from headwater catchments in Finland (Valkea-Kotinen (FI01) and Hietajarvi (FI03)) and Sweden (Aneboda (SE14), Kindla (SE15), and Gammtratten (SE16)) were provided by ICP IM (Kleemola and Forsius, 2018). Details on the catchments are given in Vuorenmaa et al. (2018).

2.4.3 Rivers

Two sets of data have been used to search for predictors of Hg concentrations in rivers. The first consists of a limited number of samples recently analysed (2018) with a sensitive Hg method (limit of quantification, LOQ, 0.2 ng/L), while the other includes a larger number of samples analysed using a less sensitive Hg method (LOQ 1 ng/L).

The Norwegian River Monitoring Programme is part of a joint monitoring programme under the “*OSPAR Commission for the Protection of the Marine Environment of the North-East Atlantic*”, where the purpose is to estimate the total loads of selected pollutants to Convention waters on an annual basis. As part of this programme, Hg is measured in rivers at different intervals and for different locations since 1990. However, there is a high analytical uncertainty related to the Hg data in the programme, following the changes of both laboratories and analytical methods between 1990 and 2015. The LOD for the program has been 1 ng/L since 1993, and since a very high proportion of the observations is and have been below LOQ, no meaningful trend assessment of the annual loads is possible (Skarbøvik et al., 2017).

Prior to 2015, two loads were estimated for each parameter in the monitored rivers: one assuming all LOQ values were zero, and another assuming all LOQ values were equal to the LOQ itself. These two estimates are reported separately as “low average” and “upper average”. Often, the difference between the two estimates is large because a large proportion of the Hg values are reported as the LOQ. We have chosen to present the average of the low and upper average when presenting data from RID collected prior to 2015. Since 2015, a new method has been applied, where the LOQ values are linearly adjusted according to the relative number of LOQ readings each year (see Skarbøvik et al. (2017) for details).

The data evaluated is for the period 2011-2014 based on monthly measurements of Hg in 10 rivers (11 in 2014); quarterly measurements of Hg in 36 rivers; and loads estimations for 109 rivers based on older data (1990-2003). For 2015 and 2016 data is based on monthly measurements of Hg in 11 rivers and quarterly measurements of Hg in 36 rivers. For 2017 data is based on monthly measurements of Hg in 20 rivers.

In 2018, NIVA’s SIS “*Land-Ocean Interactions*” has funded a project testing a more sensitive analytical method for determination of Hg in rivers. The method is described in detail elsewhere (Braaten et al., 2014a; Braaten et al., 2014b), but in short, it follows USEPA Method 1631 for determining Hg in water by oxidation, purge, and trap and cold vapor atomic fluorescence spectrometry (CVAFS) (USEPA, 2002).

2.5 Modelling of headwater Hg export

Total Hg export from Norwegian headwaters was estimated from total export of TOC, which again was estimated based on TOC input to the headwaters. A simplified model was developed which is described here. The TOC headwater inputs were estimated using land cover and runoff for all headwater catchments in Norway (Regine dataset) (Skarbøvik et al. 2017), and an equation linking TOC export to runoff, forest cover, and peatland cover (based on long-term averages of TOC export for eight

Norwegian catchments) (Garmo et al. 2015). The TOC fluxes were multiplied with a Hg-to-TOC ratio from lake and river monitoring.

The empirical model was, in short, derived in the following way: The long-term annual means of TOC export for Norwegian headwater catchments were based on multi-decadal monitoring data (except for SVART01, **Figure 2**), suggesting that these long-term averages of TOC export present a robust estimate for each catchment. Assuming that between-catchment differences in TOC export is primarily driven by differences in discharge, TOC export normalized with regard to discharge results in a 'flow-weighted' TOC discharge (in $\text{g C m}^{-2} \text{mm}^{-1}$) which is expected to be largely a function of land cover characteristics. The headwater catchments include a considerable variation in land cover, i.e. 4 to 90% forest, and 7 to 22% peatlands, 1-15% lakes, and 3-83% with little soil cover and/or vegetation.

A simple linear regression model using %forest and %peatland explained 90% of the variation in TOC Q^{-1} (**Equation 1**):

$$\text{TOC export (g C m}^{-2} \text{mm}^{-1}) = -0.308 + 0.058*\% \text{forest} + 0.290*\% \text{peatland} \quad (1)$$

The coefficients for forest and peatlands were significant ($p < 0.05$ and $p < 0.005$), while the intercept was not significant.

3 Results and discussion

3.1 Atmospheric deposition of Hg to Norway

3.1.1 Total deposition

The mean total deposition flux for mainland Norway (i.e. excluding Svalbard) in 2016 was $9.53 \mu\text{g m}^{-2} \text{ year}^{-1}$ (**Table 2**), slightly lower than the European average in 2014 ($10 \mu\text{g m}^{-2}$) (EMEP, 2016). Generally, deposition fluxes were highest in the drainage basins North Sea ($12.52 \mu\text{g m}^{-2} \text{ year}^{-1}$) and Norwegian Sea ($11.15 \mu\text{g m}^{-2} \text{ year}^{-1}$) (**Figure 3**), indicating the large amount of wet deposition of Hg in these typically wet regions of Norway. The drier regions of Norway, drainage basins Skagerrak and Barents Sea, have similar dry depositions of Hg compared to North and Norwegian Sea, but lower wet deposition.

Table 2 Deposition of Hg to mainland Norway for 2016, based on data from EMEP. Shown are mean deposition flux ($\mu\text{g m}^{-2} \text{ year}^{-1}$) and total deposition (kg year^{-1}) per main drainage basin as indicated in **Figure 2**. N/A is the geographical area of Norway that drains towards Sweden.

Drainage basin	Land area (km ²)	Mean total mercury deposition (kg year ⁻¹) and flux ($\mu\text{g m}^{-2} \text{ year}^{-1}$)		
		Wet deposition	Dry deposition	Total deposition
Skagerrak	98695	553 5.60	194 1.96	747 7.56
North Sea	47034	493 10.49	96 2.03	589 12.52
Norwegian Sea	80983	771 9.51	132 1.64	903 11.15
Barents Sea	82165	589 7.17	143 1.74	732 8.91
N/A	13997	82 5.89	24 1.69	106 7.58
All of mainland Norway	322874	2488 7.71	589 1.82	3077 9.53

Looking at literature for deposition of total Hg to Norway, a few studies exist for Scandinavia that are suitable for comparison. Iverfeldt (1991) documented a south-to-north gradient of decreasing wet Hg deposition, ranging from 27 to $3 \mu\text{g m}^{-2} \text{ year}^{-1}$. Similar values were obtained by Munthe et al. (1998) when comparing modelled (2 - $15 \mu\text{g m}^{-2} \text{ year}^{-1}$) and measured (5 - $27 \mu\text{g m}^{-2} \text{ year}^{-1}$) wet Hg deposition to four Swedish stations between 1988 and 1990. Our calculated total annual Hg deposition flux for 2016 falls within the lower range of these previous estimates which is as expected considering the reduced atmospheric Hg concentrations in Europe. However, the spatial resolution of our data is probably too low to find similar north to south gradients as reported elsewhere. It is clear that the variation in Hg deposition is partially governed by precipitation patterns.

Total Hg deposition for Norway, referred to by Munthe et al. (2007), was $4349 \text{ kg year}^{-1}$ (year not given), which is considerably higher than the estimate we report, i.e. 3077 kg for 2016. Inter-annual

variation might be considerable because of variation in precipitation. Additionally, the reported decline in concentrations and deposition of Hg in Europe may account for some of the difference between the value reported here and the value from Munthe et al. (2007).

Other studies looking at only the wet Hg deposition fluxes to Scandinavia during the 1990s also show values similar to our findings. Examples include a study from a Finnish site measuring $5.7 \mu\text{g m}^{-2} \text{ year}^{-1}$ Hg in wet deposition in 1994 and 1995 (Porvari and Verta, 2003). From a research station south in Norway (Lista), Berg et al. (2006) reported wet deposition of Hg from 9 to $21 \mu\text{g m}^{-2} \text{ year}^{-1}$ between 1990 and 2002, while Munthe et al. (2001) reported wet deposition between 2 and $25 \mu\text{g m}^{-2} \text{ year}^{-1}$ for four stations throughout Sweden between 1987 and 1997. Lee et al. (1998) documented atmospheric inputs of Hg including both open field deposition and litterfall from approximately 20 to $60 \mu\text{g m}^{-2} \text{ year}^{-1}$ for three sites in Sweden and Finland.

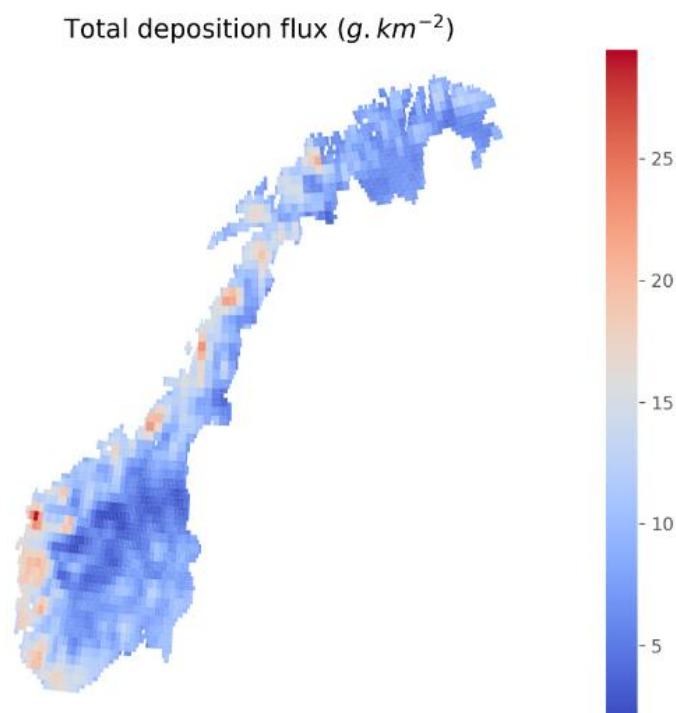


Figure 2 Total deposition flux (g km^{-2}) of Hg to mainland Norway in 2016, based on data from EMEP.

3.1.2 Deposition to different land cover

Total Hg deposition to the various land cover types varies mainly depending on the total area for each land cover type (**Figure 4**). Open areas received $1\,827 \text{ kg Hg}$ and constitute 56% of the Norwegian land (e.g. grassland, wetland/peatland, cropland, snow/ice, shrublands and barren lands), while the remaining Hg was distributed as 946 kg to forest (37% of land cover), 251 kg to water (7%), and 7 kg to developed areas (0.2%).

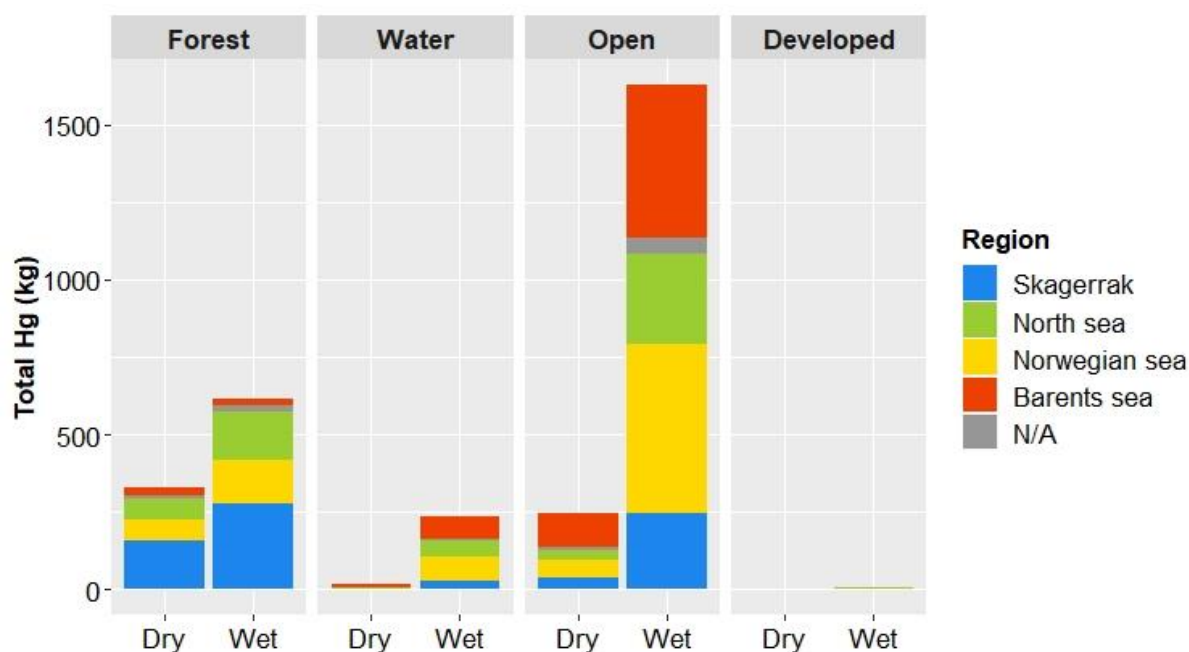


Figure 3 Sum of dry and wet Hg deposition (kg) to the varies land cover types forest, water, open areas, and developed land for each of the catchment regions.

For all land cover types (i.e. forests, open areas, waters, and developed land), wet deposition was much higher than dry deposition (**Figure 4**). For open areas, waters, and developed land, wet deposition was from 6 to 16 times higher than dry deposition, while for forests the wet deposition was only twice that of dry deposition. This reflects the importance of vegetation for scavenging of Hg from the atmosphere. Because of the large surface area of vegetation (e.g. leaves etc.), dry deposition is typically much more significant in forested areas than in open areas (Graydon et al., 2012; Miller et al., 2005; Risch et al., 2012).

The importance of dry deposition to forests is also evident when looking at fluxes of Hg deposition (**Table 3**), which is independent of land cover prevalence. While the wet deposition flux is almost identical across all land cover types ($7.8 - 7.9 \text{ g km}^{-2} \text{ yr}^{-1}$), the dry deposition to forested sites is 5-11 times higher than to the other land covers. This suggests that lakes in forested catchments are prone to a higher Hg loading from air when compared to the other land cover types. Also, forested sites typically have soils rich in OM which is associated with a high Hg storage capability (Obrist et al., 2016; Yu et al., 2014). The stored Hg can subsequently be released and transported to the lakes in association with dissolved OM (DOM) (Åkerblom et al., 2008).

Table 3 Mean dry and wet Hg deposition flux ($\mu\text{g m}^{-2} \text{yr}^{-1}$) to the various land cover types for each of the main drainage basins.

Drainage basin	Dry and wet mercury deposition fluxes to different land covers ($\mu\text{g m}^{-2} \text{year}^{-1}$)							
	Forests		Waters		Open areas		Developed land	
	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
Skagerrak	3.4	5.5	0.2	5.8	0.8	5.6	0.4	5.8
North Sea	4.9	10.7	0.3	10.7	1.0	10.6	0.6	11.6
Norwegian Sea	5.3	9.8	0.4	9.9	1.0	9.7	0.6	8.8
Barents Sea	10.0	7.3	0.9	7.0	1.8	7.1	2.8	7.8
N/A	4.7	5.8	0.5	6.1	1.1	6.0	0.4	5.0
All of mainland Norway	5.7	7.8	0.5	7.9	1.1	7.8	1.0	7.8

3.1.3 Hg in throughfall and litterfall

Total Hg deposition to Norwegian forests was estimated based on the sum of litterfall and throughfall. This was done by combining measured Hg levels in litterfall and throughfall from four extensively studied forested sites (ICP IM program) with an estimate of total litterfall and throughfall in Norwegian forests. First, an evaluation of the findings from the case studies will be presented, and followed by a presentation of the final estimates of total Hg deposition to Norwegian forests.

In **Table 4**, concentration and fluxes of Hg measured in litterfall, throughfall, and precipitation at the four case study sites are presented. Note that the number of years with observations varied between locations and type of flux data. Total annual Hg deposition was found to be one to two times higher in the spruce forests (Aneboda $27.5 \mu\text{g m}^{-2}$, Kindla $22.4 \mu\text{g m}^{-2}$, and Langtjern $20.4 \mu\text{g m}^{-2}$) than in the pine forest site at (Langtjern $10.4 \mu\text{g m}^{-2}$). Note that throughfall data was not available for the Gammtratten site. Relative contribution of Hg litterfall and throughfall was also found to differ between the two types of forests: In the spruce forests, Hg litterfall commonly exceeded that of throughfall, while for the pine forest site the opposite was the case (**Table 4**). This particular pine forest had a very low density which likely result in low interception by the canopy and correspondingly low litterfall (which is a function of needle biomass).

In the spruce forests, both the Hg litterfall flux and concentration displayed a decreasing south-to-north trend (flux: $19.0 - 6.8 \mu\text{g m}^{-2}$ and concentration: $77 - 50 \text{ ng g}^{-1}$) (**Table 4**). This suggests that interception of Hg by needles declines towards the north, which is possibly related to lower atmospheric concentration of Hg. A similar trend was not found in the data from EMEP (section 3.1.1), but has previously been described in the literature for deposition (Munthe et al., 2007). For throughfall and precipitation, there was a consistent spatial pattern similar to that of litterfall. However, the Hg flux in throughfall did not match the concentration pattern. This is because the amount of throughfall was a stronger driver of Hg flux than concentration.

Table 4 An overview of forest Hg fluxes and concentrations from small, intensively monitored forested catchments in Norway and Sweden. Sites Aneboda, Kindla, and Gammtratten refer to ICP IM monitoring sites SE14, SE15, and SE16, respectively. Langtjern is also part of ICP IM (NO03)

Specifications	Site (latitude)				
	Aneboda ^a (57.07)	Kindla ^a (59.45)	Langtjern (60.22)	Langtjern (60.22)	Gammtratten ^a (63.51)
Forest type	Spruce	spruce	spruce	pine	spruce
Litterfall (LF)					
$\mu\text{g Hg m}^{-2} \text{ yr}^{-1}$	19	12.3	9.9 ^b	2.7 ^b	6.8
$\mu\text{g m}^{-2} \text{ yr}^{-1}$	272	159	178 ^b	97 ^b	123
ng g^{-1}	72.2	77.1	50.2 ^b	30.3 ^b	56.1
Throughfall (TF)					
$\mu\text{g Hg m}^{-2} \text{ yr}^{-1}$	8.5	10.1	9.5 ^{b, c}	7.7 ^{b, c}	n.d.
mm	658	604	-	-	-
ng L^{-1}	15.9	16.6	12.8 ^{b, c}	10.4 ^{b, c}	-
Precipitation (P)					
ng L^{-1}	6.6	4.7	4.5 ^{b, c}	4.1 ^{b, c}	2.2
Years of data					
LF/TF/P	8/2/2	8/2/2	1/2/1	1/2/1	8/2/-

Sources of data: ^a Kleemola and Forsius (2018); ^b Larssen et al. (2008); ^c de Wit et al. (unpublished).

For the upscaling from case sites to the whole of Norway, the mean litterfall concentration at the three southernmost sites (67 ng g^{-1}) was assumed to be representative for the overall Norway, except from the Barents Sea drainage basin, for which the value from the northernmost most site was used (56 ng g^{-1}) (Table 5).

The mean forest Hg deposition in Norway, calculated from estimated litterfall and throughfall, multiplied with measured Hg concentrations in litterfall and throughfall, adds up to $5.6 \mu\text{g m}^{-2} \text{ yr}^{-1}$ (the sum of litterfall ($2.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$) and throughfall ($3.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$). When multiplied with forest area: 678 kg Hg) (Table 5). These values are lower than what is modelled from EMEP, where forest Hg deposition in Norway is estimated to be $13.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$ or 946 kg (Table 3, Figure 4). Note that we did not include estimates of Hg inputs from throughfall to forest above the coniferous forest limit (NFI region 8, Table 5). This is mostly birch forest with low density that covers a relatively large area. The contribution from this region to the total Norway budget is potentially large as it covers a large portion of Norway, but the quality of the data is poor. Hence, it adds too much uncertainty to include it in the calculations: e.g. if using the pine concentrations obtained above, the throughfall inputs are doubled.

The sum of litterfall and throughfall in the Nordic case studies (i.e. 27, 22 and $21 \mu\text{g m}^{-2}$ in spruce forest; $10 \mu\text{g m}^{-2}$ in pine forest, Table 4) is higher than the EMEP and the Norwegian forest sum of litterfall and throughfall estimate (Table 5). Whether the range in estimates from the three different methods is related to lack of representativity (Nordic case studies) or limitations in the methodology or data availability is difficult to assess within the scope of this report.

Note that the mean total deposition for forests in Norway, obtained from EMEP, is $13.5 \mu\text{g m}^{-2}$ and lower than for site-specific results for spruce forest but higher than for pine forest. Additionally, the

south to north decline in Hg concentrations in the site-specific results does not match the pattern for the four drainage basins, where deposition to the southernmost drainage basin (Skagerrak) is much lower than for Barents Sea in the north. A more extensive comparison of EMEP-modelled deposition to forests with measured deposition data would be valuable but is outside the scope of this report.

Table 5 Estimations of litterfall (LF) and throughfall (TF) Hg fluxes for different regions in Norway. Details on regions and calculations are found in de Wit et al. (2015). N/A is 'not attributed' to drainage basin.

Drainage area	NFI Region	Area 10 ³ km ²	Volume forest type				LF g m ⁻² yr ⁻¹	Hg in LF		LF Hg flux µg m ⁻² yr ⁻¹	TF factor %	TF mm	Hg in TF		TF Hg flux µg m ⁻² yr ⁻¹
			Total 10 ³ km ²	Spruce (%)	Pine (%)	Birch (%)		spruce ng g ⁻¹	pine ng g ⁻¹				spruce ng L ⁻¹	pine ng L ⁻¹	
Skagerrak	1	29.8	21.8	50	37	13	110	67	30	5.3	75	283	15	10	3.6
	2	23.8	17.4	56	27	17	110	67	30	5.6	75	291	15	10	3.8
	3	21	15.4	33	44	24	102	67	30	4.4	87	548	15	10	6.6
North Sea	4	26	13.4	29	36	35	88	67	30	3.6	87	873	15	10	10.3
Norwegian Sea	5	23.7	14.8	59	21	20	92	67	30	4.8	87	522	15	10	6.9
	6	25.4	15.4	26	11	62	46	67	30	1.8	87	561	15	10	6.5
Barents Sea	7	48.6	10.3	0	30	70	12	56	30	0.4	95	276	10	10	2.9
N/A	8	125.5	11.3	4	7	89	21	30	30	0.6	95	649	10	10	0.0
Norway total	-	323.8	37.6	-	-	-	-	-	-	2.3	-	-	-	-	3.3

3.2 Hg in headwaters

Although aqueous chemistry is documented in many streams and lakes as part of national monitoring programs in Norway, Hg is not routinely measured. Hence, data on Hg in these aqueous systems is scarce, and data availability is restricted to various research projects. However, stream water Hg data were available from the ICP IM network (Vuorenmaa et al., 2018), which are small, natural catchments with first-order streams (headwater catchments). The Norwegian lake data reported in this work are also mostly located in small headwater catchments, and for the sake of simplicity called headwater lakes here.

3.2.1 Headwater lakes

Distribution of Hg concentrations available in literature are restricted to lakes from southeast Norway and northern areas (i.e. Finnmark) while excluding the mid part of the country (**Figure 2**). Concentration distributions ranged over more than an order of magnitude (**Figure 5**), from 0.3 to 6.6 ng L⁻¹ and with a mean concentration (\pm one standard deviation) of 2.8 ± 1.8 ng L⁻¹. Typically, concentrations were higher in southern Norway compared to northern Norway, and thus, data entries were separated in two groups: lakes draining to Skagerrak (3.4 ± 1.6 ng L⁻¹) and to the Barents Sea (0.9 ± 0.7 ng L⁻¹). The Hg concentrations were significantly different in the two regions of Norway ($p < 0.0001$), as previously documented in literature (Braaten et al., 2014a). The increasing north-to-south concentration gradient for Hg is not surprising, as the same gradient is observed for TOC (Skjelkvåle et al., 1997) and also to some degree for deposition (**Figure 3**).

As expected there was a strong correlation between concentrations of Hg and TOC overall ($y = 0.34x - 0.36$; $r^2=0.79$). This is an indication of the role of organic matter as a transport vector for Hg species from catchments to surface waters (Braaten et al., 2014a; Eklof et al., 2012). Interestingly, in our data set, the linear Hg-to-TOC relationships are slightly different for the different regions (**Figure 5**). For the lakes draining to the Barents Sea, the linear regression slope (0.17) is weaker than the slope for the lakes draining to Skagerrak (0.31, ANCOVA $p=0.2$). This indicates that in the south of Norway, there is “more Hg per the organic matter”, potentially explained by: *i*) higher sulphate availability in the south compared to the north; *ii*) more forest in the south leading to higher Hg interception, higher Hg deposition and more soil accumulation; and *iii*) different OM characteristics, where more sulphur binding sites in the south is possible. Reduced sulphur sites on OM are expected to be important binding sites for Hg (Ravichandran, 2004).

The strong and significant relationship between Hg and TOC for headwaters are further utilised for calculation of catchment export and headwater export (see section 3.2.3 below).

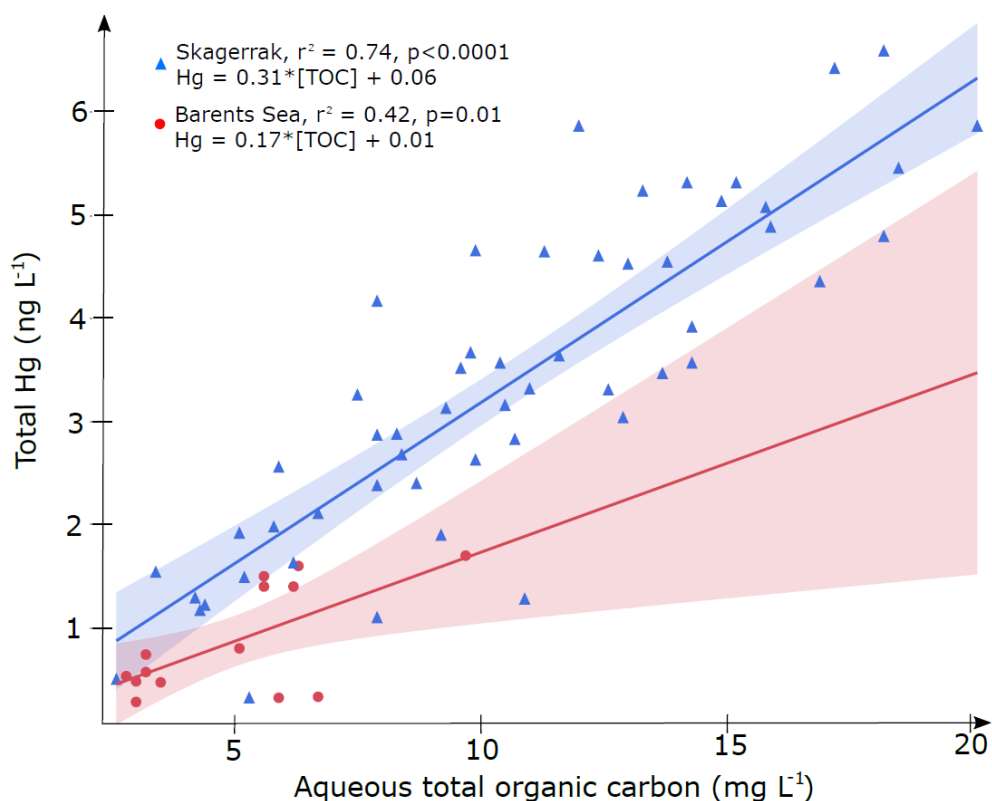


Figure 4 Concentrations of Hg (y-axes) and aqueous total organic carbon (TOC, x-axes) in 70 lakes in Norway. Lakes are from areas draining to Skagerrak (blue triangles) and the Barents Sea (red circles).

3.2.2 Streams

Stream Hg concentration data were available for eight streams at three locations (i.e. southeast Norway (Langtjern, ca. 100 km NE of Oslo, **Figure 2**), south Finland (ca. 150 km north of Helsinki) and eastern Finland (at 64°N)) (all included in the ICP Integrated Monitoring network). All sites are forested, coniferous catchments with a varying % peatland cover. The median stream Hg concentrations for the entire monitoring time series vary between 1 and 6 ng L⁻¹, while median TOC varies from 6 to 17 mg C L⁻¹. The median Hg-to-TOC ratio varies less than the separate components of the ratio, i.e. from 0.18 to 0.32, suggesting that the variation of Hg concentrations between the streams is partly driven by TOC (**Table 6**).

Based on all data, there is a significant positive relationship between Hg and TOC ($r^2 = 0.43$, $p < 0.0001$, slope 0.26). When separated per region or stream, the correlation remains significant but the slope of the relationship is significantly lower at Langtjern (0.12) than at the Finnish sites (0.29) (ANCOVA, $p < 0.0001$), indicating that TOC in Finland is loaded with more Hg than at Langtjern (**Figure 6**). The Finnish sites have a slope similar to that of lakes in the Skagerrak drainage area (**Figure 5**). Within the Langtjern catchment, the LAE11 catchment has a lower Hg-to-TOC ratio than the neighbouring catchments, suggesting a lower load of Hg per g TOC in the LAE11 catchment. However, the slopes of the Hg-to-TOC relationships are not significantly different between the four stations at Langtjern (ANCOVA, all $p > 0.05$). Note that the precision of the Hg measurement methods provide data with one decimal in the Langtjern dataset and zero decimals in the Finnish dataset.

Table 6 Stream total Hg concentrations (mean, median and 25 and 75 % quartiles (0.25-q, 0.75-q)), TOC concentrations (median and quartiles), and median Hg:TOC ratio for four sites in Norway (four streams) and Finland (four streams). Number of sampling points are shown (n).

Catchment	Stream	n	Total Hg (ng L ⁻¹)				TOC (mg L ⁻¹)			Hg:TOC
			mean	median	0.25-q	0.75-q	median	0.25-q	0.75-q	median
Langtjern	LAE01	119	3.5	2.8	0.8	3.9	11.5	9.9	12.7	0.32
	LAE02	115	3.3	2.8	0.8	3.5	11.5	10.0	13.6	0.27
	LAE03	144	3.2	3.6	1.1	3.6	11.8	10.1	14.8	0.24
LAE11	LAE11	147	3.9	4.7	1.3	4.4	17.4	14.9	21.4	0.21
Valkea-Kotinen	FI01-1	203	6.4	6	3	7	20.1	17.7	23.8	0.26
	FI01-3	108	6.1	5	4	7	14.4	13.0	16.5	0.31
Hietajarvi	FI03-1	204	1.6	1	1	2	6.1	5.3	6.9	0.18
	FI03-7	203	2.5	2	1	3	11.0	9.3	14.0	0.18

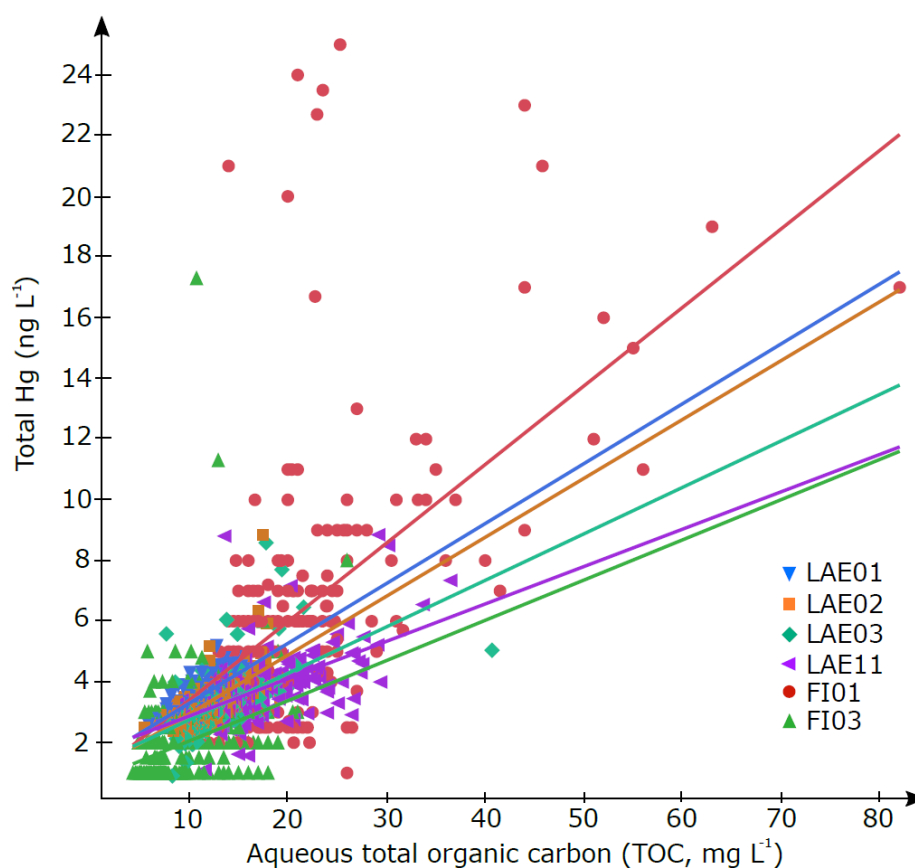


Figure 5 Total Hg (ng L⁻¹) versus TOC (mg L⁻¹) in 4 streams at Langtjern (outlet (LAE01), and two inlets (LAE02 and LAE03), in addition to LAE11 stream from neighbouring catchment) and two Finnish ICP IM sites (FI01 and FI03). Sampling period is from 2004 to 2017 (n=1 020). All regression lines are significant ($p < 0.0001$) with r^2 and slopes being: LAE01 ($r^2=0.46$, slope 0.20); LAE02 ($r^2=0.39$, slope 0.19); LAE03 ($r^2=0.34$, slope 0.15); LAE11 ($r^2=0.29$, slope 0.12); FI01 ($r^2=0.29$, slope 0.26); and FI03 ($r^2=0.11$, slope 0.13).

The slopes for the Hg-to-TOC relationships are not completely similar in the streams (**Figure 6**) compared to what was found for the headwater lakes (**Figure 5**). For the lakes, our data show a difference in slopes between the North (i.e. Barents, 0.17) and the South (i.e. Skagerrak, 0.31), while the streams in the Langtjern catchments have lower slopes than what was found for the lakes in the south. At Langtjern, both the lake outlet (LAE01, 0.20) and the two inlets (LAE02, 0.19, and LAE03, 0.20) had slopes similar to what was found in the Northern lakes (0.17). This is surprising, as we were expecting lake outlets to have similar slopes to that of headwater lakes. However, the deposition to the Langtjern catchments was lower than other sites (throughfall and litterfall in **Table 3**). In combination with potentially higher TOC from more peatlands compared to other south-Norwegian sites, this could be an explanation for the lower slopes at Langtjern.

3.2.3 Headwater Hg export

To evaluate the role of aquatic removal of Hg between headwaters and what is exported to the coast, total export of Hg from headwaters was derived from the total TOC input to headwaters, which here is equivalent to catchment export of TOC. It is well-known that TOC export is a function of land cover and climate (Larssen et al. 2011). However, no model was available for estimating total TOC headwater export. Using the Norwegian headwater data, we derived a simple statistical model for long-term TOC export (see method section) and used data on land cover and discharge (Regine-watershed data) to upscale to Norway (>20 000 headwater catchments) (see a description of the Teotil model in Skarbøvik et al., 2017).

For all ICP IM catchments, discharge, TOC, and Hg data were available allowing the calculation of element fluxes, which are presented in **Table 7**. Note that the periods for which data were available vary between sites (**Table 7**). Additionally, note the difference in mean annual runoff for the sites, i.e. around 200 mm at Valkea-Kotinen (FI01) and over 700 mm at Langtjern.

The mean annual fluxes of Hg (**Table 7**) varied with a factor of 4, from 0.6 $\mu\text{g m}^{-2}$ in Hietajarvi (FI03) to 2.8 $\mu\text{g m}^{-2}$ in Langtjern. To evaluate possible factors explaining the between-site differences, Hg export was normalized with regards to TOC export and discharge (**Table 8**). When normalized for TOC, the differences between the sites were reduced, i.e. varied with less than a factor 2 (0.22 – 0.37). On the other hand, when normalized for discharge, the differences between the sites were not reduced (1.6-6.4). Thus, when considering TOC export, part of the inter-site variation in Hg export can be explained by TOC. The remaining variation could be related to Hg deposition, which may be related to litterfall (related to forest density) and precipitation inputs (clearly contrasting between the sites). We conclude that the most important factor that explains spatial variation in Hg export between sites is TOC.

Table 7 Fluxes of mercury (Hg), total organic carbon (TOC) and water (discharge, Q) in three headwater sites. Data is based on monthly sampling.

Catchment	Name	LANGTJERN				VALKEA-KOTINEN		HIETAJARVI	
	ID	LAE01	LAE02	LAE03	LAE11	FI01-1	FI01-3	FI03-1	FI03-7
Catchment area	km ²	4.8	n.a.	0.8	0.3	0.2 ^b	n.a.	4.6	0.8
TOC	g C m ⁻² yr ⁻¹	7.8	8.6	9.1	12.7	4.1	3.4	2.3	3.8
	Years of data	2009-2017	2009-2017	2009-2017	2009-2017	2000-2015	2000-2008	2000-2015	2000-2015
Hg	µg Hg m ⁻² yr ⁻¹	2.5	2.5	2.4	2.8	1.2	1.3	0.6	0.8
	Years of data	2009-2017	2009-2017	2009-2017	2009-2017	2003-2015	2003-2008	2006-2015	2006-2015
Q	mm	739	748	748	748	188	217	370	302
	Years of data	2009-2017	2009-2017	2009-2017	2009-2017	2000-2015	2000-2008	2000-2015	2000-2015 ^a

^a No 2011 data exist for discharge (Q) at site FI01-7. ^b Vuorenmaa et al. (2014).

Table 8 Mercury (Hg) data from **Table 7** normalized with regards to total organic carbon (TOC) and discharge (Q).

Specification	Unit	LAE01	LAE02	LAE03	LAE11	FI01-1	FI01-3	FI03-1	FI01-7
Hg/TOC	µg g ⁻¹	0.33	0.29	0.27	0.22	0.29	0.37	0.25	0.22
Hg/Q	ng mm ⁻¹	3.4	3.4	3.3	3.8	6.4	5.9	1.6	2.7
TOC/Q	mg mm ⁻¹	10.6	11.5	12.2	16.9	22.0	15.8	6.3	12.5

The statistical model used to calculate TOC export for all Norwegian headwaters is based on data from long-term records in 8 Norwegian headwater catchments (**Table 9**), and more details are given in the methods section. The model uses %forest and %peatlands, and multiplies with discharge (normalized 1960-1990) to estimate TOC export, and is summed for all headwaters in a given drainage basin (**Table 10**). The TOC export in the drainage basins varied between 3.4 and 4.7 g C m⁻² year⁻¹ (**Table 10**), which is lower than the site median for Norwegian headwaters (**Table 9**). It should be noted that headwaters without forest and peatlands have a TOC export of zero, according to this model (all negative export values, related to the negative intercept, were set at zero). Large parts of Norway consist of low-density forest and tundra/shrub vegetation. Those areas contribute to TOC export, but may not be classified as 'forest' in the land cover maps. Thus, a model that only consists of peatland and forest land cover will likely underestimate landscape TOC export, and is also likely to underestimate Hg export. Thus, the estimates of drainage basin TOC and Hg export have considerable uncertainty.

Hg headwater export

For the Norwegian headwater catchments, Hg has only been measured at Langtjern, while the other stations are described by discharge and TOC (Garmo et al., 2015). Therefore, the Hg-to-TOC relationships previously established for lakes and streams in this report were used to calculate Hg export from the headwaters. We assumed a Hg-to-TOC ratio of 0.2 for the far north of Norway (Barents Sea drainage basin) and a ratio of 0.3 for the rest of Norway (**Table 9**). This resulted in a Hg export flux

that varied between 0.3 and 3.8 $\mu\text{g m}^{-2} \text{ year}^{-1}$, and with a median for all sites at 1.8 $\mu\text{g m}^{-2} \text{ year}^{-1}$. This constitutes a somewhat broader range than what was empirically estimated for the ICP IM catchments, i.e. 0.6-2.8 $\mu\text{g Hg m}^{-2} \text{ year}^{-1}$ (Table 8).

The total headwater Hg export for Norway, including all these assumptions, is estimated to be 295 kg per year. A quantitative uncertainty estimate is not possible, but given the likely underestimation of the TOC export as explained above, and the relatively consistent Hg-to-TOC ratios, we suggest that the headwater Hg export could be twice as high as estimated here.

Table 9 Estimated annual mean Hg export in Norwegian headwater catchments. Standard deviation is based on annual means.

Station Code	Nr of yrs	Hg ($\mu\text{g m}^{-2}$)		TOC (g C m^{-2})		Hg:TOC ratio ($\mu\text{g g}^{-1}$)
		mean	stdev	mean	stdev	
BIE01	33	2.0	0.6	6.8	2.1	0.3
DALELV	29	0.3	0.1	1.7	0.4	0.2
KAE01	32	0.5	0.1	1.8	0.3	0.3
LAE01	33	1.9	0.6	6.5	1.9	0.3
LAE03	30	2.2	0.6	7.4	2.0	0.3
OVELV 19 23	25	1.1	0.3	3.6	1.0	0.3
STE01	33	1.6	0.5	5.4	1.6	0.3
SVART01	12	3.8	1.1	12.8	3.7	0.3
Site median		1.8		6.0		

Table 10 Drainage basin characteristics and estimated annual mean Hg and TOC export in Norwegian drainage basins. Nr of catchments refers to Regine headwater catchments. N/A refers to 'not attributed to drainage basin'.

Drainage basin	Nr. of Regine catchments	Total area	Area with data	Discharge	TOC export		Hg export	
	n	10^3 km^{-2}	10^3 km^{-2}	mm	g C m^{-2}	10^9 g C yr^{-1}	$\mu\text{g m}^{-2}$	kg Hg
Skagerrak	4035	99	81	803	3.4	336	1.0	101
North Sea	6454	47	40	2138	3.8	179	1.1	54
Norwegian Sea	5943	81	86	1357	4.7	381	1.4	114
Barents Sea	5655	82	93	709	2.6	214	0.5	43
N/A	876	14	0	1453	3.8	53	1.1	16
All of mainland Norway	22087	323	300	-	3.3	1055	0.9	295

3.3 Riverine export of Hg (coastal inputs of Hg)

Riverine Hg is measured through the Norwegian River Monitoring Program, but existing concentration data were found to be associated with rather large uncertainties and of poor analytical sensitivity. Therefore, an alternative way of estimating total riverine Hg export from land to the coast was undertaken: based on the more extensive and high quality TOC data available for Norwegian rivers,

established relationships between riverine Hg and TOC were used to estimate total Hg export. Here, first 1) an evaluation of the relation between riverine Hg and TOC is presented, including the RID Hg data, followed by a presentation of the estimate of total Hg export using 2) the TOC-Hg method, and 3) from measured river Hg data.

3.3.1 Relationships between TOC and Hg in Norwegian rivers

When all data available on Hg and TOC from the Norwegian River Monitoring Programme is combined (1993-2017, **Figure 7**), no significant correlation between the two variables is observed ($r^2 < 0.01$, $p = 0.49$). The lack of a significant correlation between Hg and TOC is surprising and puzzling as organic matter is a common transport vector for Hg in catchment streams (Eklof et al., 2012). Although Hg-to-TOC relationships are less frequently reported/observed for larger rivers, one would expect a relatively strong relationship (e.g. from North America: $r^2 = 0.42$, Dennis et al. (2005)). However, in the River Monitoring Programme, given the large number of Hg measurements below LOQ (1.0 ng L^{-1}), the issue is not discussed in detail. Here, we explore some of the potential reasons for the lack of correlation and discuss the effect on data evaluation.

A potential explanation could be related to the samples with high Hg concentrations ($> 20 \text{ ng L}^{-1}$), but relatively low TOC concentrations ($< 6 \text{ mg L}^{-1}$). In these samples, total suspended material (TSM) is also very low (close to 0, data not shown), so the high concentrations are not related to high particle content. Concentrations of Hg higher than 10 ng L^{-1} is not commonly observed in Norwegian aqueous environments, and a potential explanation could be contaminated samples, likely related to sampling equipment rather than Hg releases to the environment.

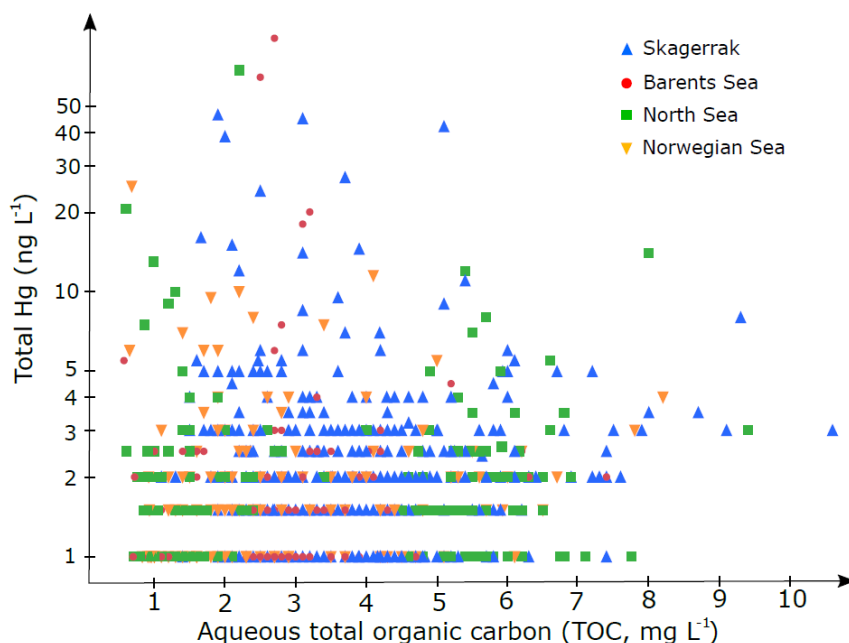


Figure 6 River concentrations of Hg (y-axis) and aqueous total organic carbon (TOC, x-axis) from the Norwegian River Monitoring Programme (1991-2017). No significant Hg versus TOC relationships are found for the drainage regions separately (all $p > 0.05$) or for all available samples combined ($n = 861$, $r^2 < 0.01$, $p = 0.49$). The y-axis is log-transformed to show the high variation in Hg concentrations.

By excluding samples with high concentrations ($\text{Hg} > 10 \text{ ng L}^{-1}$, $n=31$ samples), there is still no strong correlation observed between Hg and TOC (**Figure 8**). When samples are separated by the different drainage regions, a weak relationship is found between Hg and TOC in rivers draining to Skagerrak ($r^2 = 0.02$, $p < 0.001$). One obvious issue with the low-concentration Hg data is the precision of the analytical Hg method, resulting in reporting of only “half numbers” of Hg. Additionally, the sensitivity of the method means that all samples with $\text{Hg} < \text{LOQ}$ (1 ng L^{-1}) are excluded. This is probably why the data is unable to capture the natural variation in the samples, and why a significant Hg-to-TOC relationship is generally not observed.

A newer dataset from 2018 contains data for 11 of the 20 rivers in the current River Monitoring Programme (Alna, Drammenselva, Glomma, Skienselva, Storelva, and Otra draining to Skagerrak; Vosso draining to the North Sea, Orkla draining to the Norwegian Sea, and Målselv, Tana, and Alta draining to the Barents Sea, **Figure 2**). In the 2018 dataset (data available for January-September), 56% of the variation in the Hg concentrations can be explained by TOC ($r^2=0.56$, $p < 0.0001$), and the slope of the Hg-to-TOC relationship is 0.28. Here, it is worth noting that the variation in Hg and TOC concentrations is remarkably lower than in the old River Monitoring Programme dataset, ranging from 0.2 to 2.9 ng L^{-1} and from 0.7 to 6.7 mg L^{-1} .

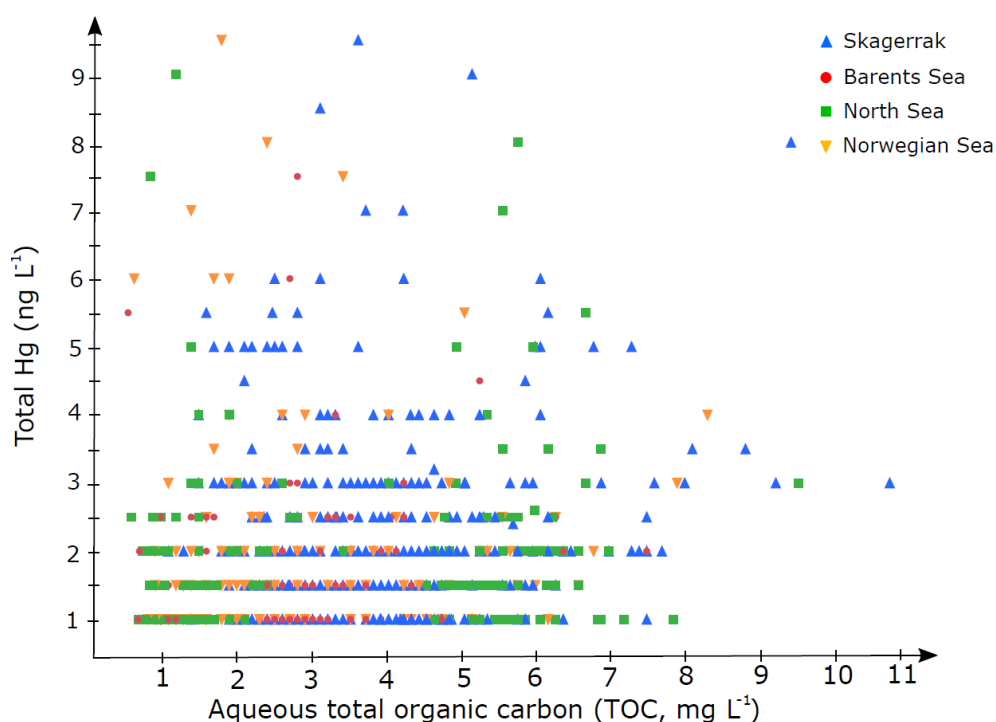


Figure 7 River concentrations of Hg (y-axes) and aqueous total organic carbon (TOC, x-axes) from the Norwegian River Monitoring Programme (1991-2017) where samples of Hg concentrations $> 10 \text{ ng L}^{-1}$ are removed. No significant Hg versus TOC relationships are found for the drainage regions Barents Sea, North Sea, or Norwegian Sea (all $p > 0.05$). A weak significant relationship is found for the Skagerrak region ($\text{Hg} = 0.15 \cdot [\text{TOC}] + 1.54$, $r^2 = 0.02$, $p < 0.001$).

The dataset from 2018 is not representing a full year of data, and some of the annual variation in Hg and TOC concentrations might be lost. However, the data enables a drainage region-specific evaluation of the Hg-to-TOC relationships (**Figure 9**), where significant relationships are found for Skagerrak ($r^2=0.61$, $p < 0.0001$), the Barents Sea ($r^2=0.50$, $p < 0.0001$), and the Norwegian Sea ($r^2=0.63$, $p=0.01$). For

the regions draining to the North Sea (no significant relationship, $r^2=0.36$, $p=0.09$) and to the Norwegian Sea, data is only from one river, hence representing temporal variation rather than spatial variation.

For the 2018 river dataset, the Hg-versus-TOC linear regression slopes are significantly different (ANCOVA, $p=0.018$) for the Skagerrak (0.35) and Barents Sea (0.19). They also show great similarity to the regression slopes found for Norwegian headwaters (**Figure 5**): 0.31 for Skagerrak and 0.17 for Barents Sea. If we group data from rivers draining to Skagerrak, the North Sea, and the Norwegian Sea, given that data from the North Sea and the Norwegian Sea only represent one river each, the regression slope is 0.27.

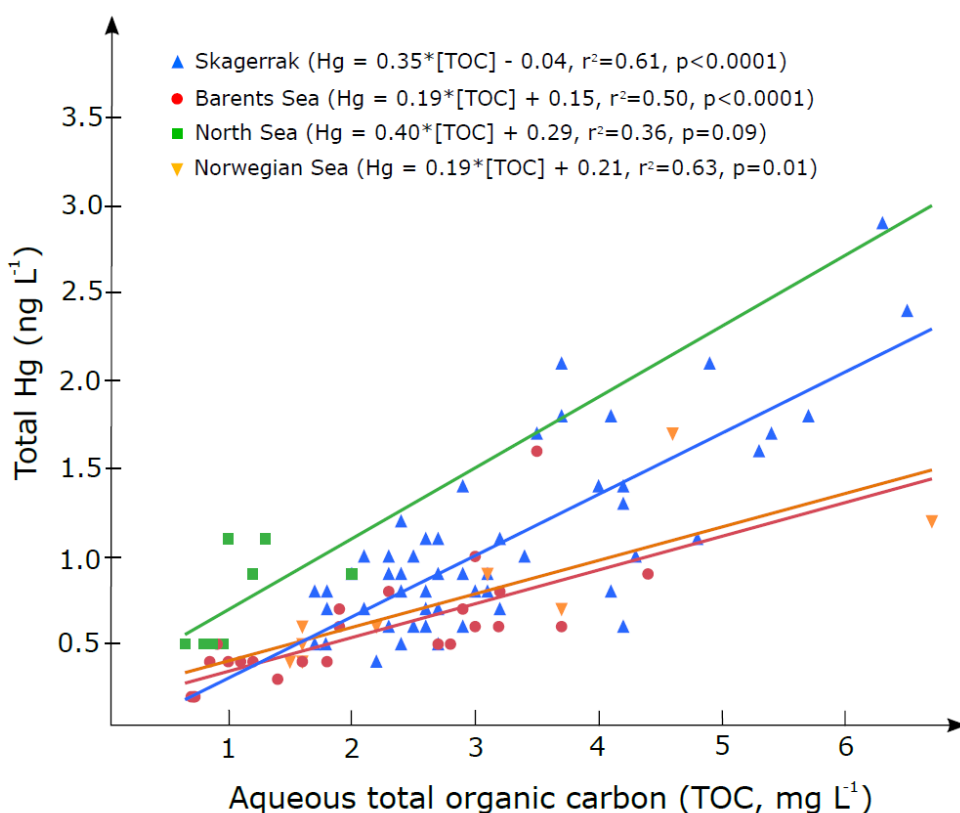


Figure 8 River concentrations of Hg (y-axes) and aqueous total organic carbon (TOC, x-axes) from 2018 (January-September). Significant Hg versus TOC relationships are found for the drainage regions Skagerrak, Barents Sea, and Norwegian Sea (all $p<0.05$).

3.3.2 Estimations of Hg export based on riverine TOC

The total riverine Hg export was estimated by relating the Hg-to-TOC-ratios to previous estimates of riverine TOC inputs to the Norwegian coast (de Wit et al., 2015). The total riverine transport presented in De Wit et al. (2015) is based on water chemistry and discharge from 46 major river outlets in Norway for 1999 to 2008, monitored under the previous (< 2017) river monitoring programme (details in e.g. (Skarbøvik et al., 2017)). Details on the methods are given in De Wit et al. (2015). In short, the monitored river catchments cover 48% of Norway, which requires upscaling to obtain an estimate of TOC export for entire Norway. Temporal extrapolation was needed to provide an annual mean TOC export for 1990-2008, the time-period for the study. Discharge was modelled with the spatially

distributed version of the hydrological HBV-model (Beldring et al., 2003). Concentrations of TOC were assumed to be equal to dissolved organic carbon (DOC). For each river, annual flow-weighted concentrations of TOC (FWC TOC) for the years 1999-2008 were calculated by dividing annual TOC export with annual discharge. Annual TOC export was summed from daily TOC export, calculated by multiplying daily discharge by daily TOC concentration. Mean annual FWC TOC and associated uncertainty was calculated for each river as the average and standard deviation of annual FWC TOC for 1999-2008. For each drainage area, a catchment area-weighted mean FWC TOC was calculated based on the individual rivers, and multiplied by mean annual discharge (1990-2008) to obtain riverine TOC export (1990-2008) for the entire drainage area. Uncertainty in river TOC export was calculated using the standard deviations of area-weighted mean FWC TOC and mean annual discharge. Total riverine TOC transport for Norway was obtained by summing per drainage area.

The riverine TOC export reported here was used to estimate riverine Hg export for Norway, using Hg-to-TOC ratios 0.3 for Skagerrak, North Sea, and Norwegian Sea, and 0.2 for Barents Sea. The reasoning for using these ratios is related to a quality evaluation of the available data. The lake (**Figure 5**) and streams (**Figure 6**) Hg-to-TOC data show similar patterns to the one-year river monitoring data from 2018 (**Figure 9**) with respect to the Skagerrak and Barents Sea regions. The regions Norwegian Sea and North Sea are in **Figure 9** only represented by one river each, and the Hg-to-TOC relationship is representing temporal variation rather than the spatial variation. Following this, the amount of total Hg to riverine coastal input is estimated to 281 ± 24 kg Hg yr⁻¹ or $0.9 \mu\text{m}^{-2}$ (**Table 11**).

Table 11 Key characteristics of river monitoring in four main drainage areas of Norway, and riverine carbon fluxes for the period 1990 to 2008.

Drainage area	Discharge ¹ (mm yr ⁻¹)	TOC ² (mg L ⁻¹)	TOC flux ³ (g C m ⁻² yr ⁻¹)	Hg-to-TOC ($\mu\text{g g}^{-1}$)	Hg export	
					(kg yr ⁻¹)	($\mu\text{g m}^{-2}$)
Skagerrak	720 (101)	3.8 (0.2)	2.8 (0.4)	0.3	87 (12)	0.81
North Sea	2395 (419)	1.9 (0.1)	4.5 (0.8)	0.3	63 (12)	1.33
Norwegian Sea	1312 (164)	2.4 (0.2)	3.1 (0.5)	0.3	111 (18)	0.93
Barents Sea	477 (46)	3.6 (0.5)	1.7 (0.3)	0.2	20 (2)	0.40
Norway	-	-	3.0 (0.3)	-	281 (24)	0.87

¹Discharge expressed as mean annual discharge (1990-2008) with one standard deviation in brackets; ²Flow-weighted, area-weighted mean (1999-2008) and one standard deviation in brackets; and ³TOC flux calculated from discharge with one standard deviation in brackets.

3.3.3 River monitoring data on Hg

In the long-term Norwegian River Monitoring dataset, relatively newer Hg data (2008-2017) were selected to estimate total Hg export. In **Table 12** the total load of Hg to the coastal areas from Norway is presented, and vary between 206 (2016) and 487 kg (2011). The river load is estimated based on measurements of Hg in 46 rivers (47 from 2014-2016), which have limited analytical certainty (**Figure 8**). The data was up-scaled to the whole of Norway based on river discharge (**Table 12**). Using these data, the mean riverine runoff of Hg for the period 2008 to 2017 is 331 ± 87 kg year⁻¹ (mean of the annual means in **Table 12**), equalling a total riverine loads of $1.0 \mu\text{g m}^{-2}$. The total load is relatively similar to the estimate provided from the Hg-to-TOC relationship method (281 ± 24 kg Hg yr⁻¹ or $0.9 \mu\text{m}^{-2}$). The two estimates are not based on the same monitoring period and are not upscaled using

the same method. The reasonable agreement between the two estimates gives credibility to the assessment of riverine export of Hg. However, given the limitations in the Hg concentration data, more reliable estimates of riverine Hg output would be obtained with a more sensitive analytical method for aqueous Hg than what is currently used. Such estimates would allow for a better characterization of inter-annual and long-term change in riverine Hg export.

Table 12 Data from the Norwegian River Monitoring Program, 2008-2017. Shown are mercury (Hg) river discharges for monitored rivers for each year and the mean \pm one standard deviation for all years. Shown are also the discharge for the monitored rivers and for the total of Norway, and the up-scaled river discharge of Hg for the whole of Norway.

Year	Hg river discharge (kg year ⁻¹)		River discharge (10 ⁶ m ³ day ⁻¹)	
	Monitored rivers	Total for Norway	Monitored rivers	Total for Norway
2008	196	328	592	990
2009	286	438	535	819
2010	138	235	442	752
2011	280	487	614	1068
2012	223	382	558	956
2013	199	341	531	909
2014	190	314	580	957
2015	178	313	633	1112
2016	115	206	519	928
2017	148	269	621	1129
Mean 2008-2017	195 \pm 56	331 \pm 87	563 \pm 58	962 \pm 121

3.4 The Hg budget for Norway

Total atmospheric deposition is estimated based on modelled numbers from EMEP to a total of 3077 kg or 9.5 $\mu\text{g m}^{-2} \text{yr}^{-1}$. Of this, wet deposition equals 81% (2488 kg) and dry deposition 19% (589 kg). Of the total deposition to Norway, forests, open areas, surface waters and developed areas receive 946 kg, 1872 kg, 251 kg, and 7 kg, respectively (**Figure 10**). For forests, dry deposition is a significant process, and more important than for the other regions, given the vegetation effect for atmospheric scavenging of Hg. Based on EMEP modelling, wet (617 kg) and dry deposition (329 kg) of Hg to forested areas sums to 946 kg or 13.5 $\mu\text{g m}^{-2} \text{yr}^{-1}$. This is substantially more than what we have estimated based on an upscaling of forest monitoring data on Hg in litterfall and throughfall for the whole of Norway (5.6 $\mu\text{g m}^{-2} \text{yr}^{-1}$). However, when comparing with the most densely forested part of Norway (Skagerrak drainage basin), the estimates were more similar (i.e. EMEP 9 $\mu\text{g m}^{-2}$; forest monitoring: 10 $\mu\text{g m}^{-2}$). The upscaling to Norway from forest monitoring data is uncertain for high-elevation and high-latitude areas with open, low productivity forest that gradually transitions to shrublands and tundra. Here, estimates of litterfall and throughfall are difficult to estimate. This is also underlined by the comparison with the case studies in Norway and Sweden, where the sum of throughfall and litterfall suggested higher deposition (i.e., 19.4 – 27.5 $\mu\text{g m}^{-2} \text{yr}^{-1}$) than the EMEP modelled estimates.

The forest deposition estimates are completely independent, relying on different data and assumptions. A common uncertainty for both is the difficult role of re-emission of Hg. In the current EMEP model, Hg deposition is treated as total (downward) deposition flux. Secondary and natural emissions are included in the estimation by prescribed fluxes from various surfaces as a function of environmental parameters (temperature, solar radiation etc.), but this does not include re-emission of Hg (EMEP, Oleg Travnikov, pers. comm.). However, Selin et al. (2008) defines three significant Hg re-emission processes: 1) “prompt” recycling; 2) evapotranspiration; and 3) soil volatilisation. Particularly interesting for the deposition to Norwegian forests are the so-called “prompt” recycling, defined as “a fraction of deposited Hg(II) [that] is quickly converted to Hg(0) and re-emitted”. Selin et al. (2008) has estimated that globally, 24 % of Hg deposited is re-emitted to the atmosphere as “prompt” recycling. If we compare this to our Norwegian budget, this would imply a lowering of the modelled deposition estimates. How estimates of litterfall and throughfall can be impacted by re-emission of Hg is not immediately clear. In theory, it is possible that internal cycling of Hg in the forest can lead to overestimation of deposition as the same molecules are measured several times. We conclude that the estimates from EMEP and forest monitoring are in reasonable agreement with each other, but that both estimates would benefit from validation with empirical data.

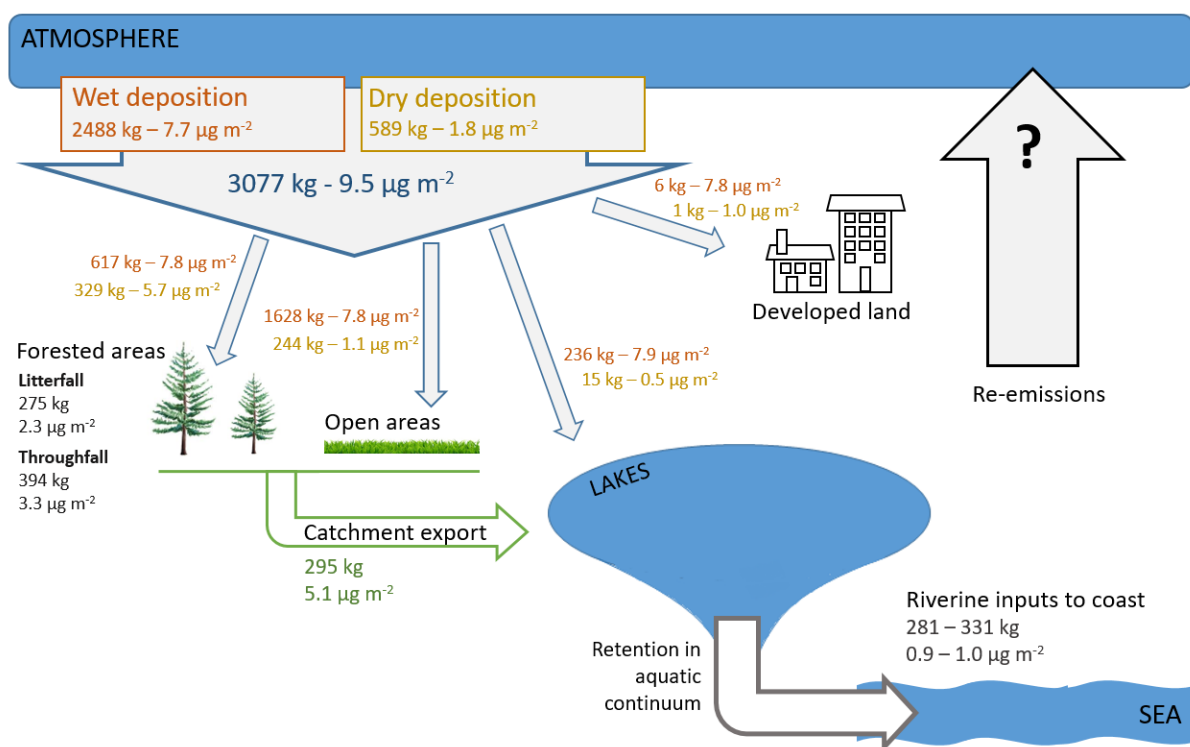


Figure 10 An environmental Hg budget for Norway showing total deposition (in kg) and deposition per unit area (in $\mu\text{g m}^{-2}$).

Regarding surface water loading, the EMEP model shows that 251 kg ($8.4 \mu\text{g m}^{-2} \text{ yr}^{-1}$) is deposited directly to surface waters from the atmosphere. We estimated catchment export to be 295 kg ($0.9 \mu\text{g m}^{-2}$), where we noted that this estimate was very uncertain and likely to be too low. Still, it is noteworthy that both estimates (catchment loading and deposition to surface waters) are in the same order of magnitude, which could imply that significant reductions in wet deposition of Hg would also

lead to significantly lower inputs of Hg to surface waters, with positive implications for Hg accumulation in the aquatic foodweb.

The Norwegian River Monitoring Programme estimates that between 2008 and 2017, 331 ± 87 kg or $1.0 \mu\text{g m}^{-2}$ are the total riverine inputs to Norwegian coastal areas annually. This is similar to what we have estimated based on a carbon budget and the significant relationships between carbon and Hg in river systems: 281 ± 24 kg Hg or $0.9 \mu\text{g m}^{-2}$. The numbers suggest that of the annually deposited Hg to Norwegian mainland from the atmosphere (3077 kg), only approximately 10 % are transported from land to the coast every year. The estimates coastal inputs of Hg are likely to be reliable relative to other flux estimates in this report because both components (i.e. runoff and concentrations) have an empirical basis. Hg-to-TOC ratios showed limited variation between rivers and lakes, suggesting that Hg and organic carbon cycling are closely linked. However, for a quantification of inter-annual and long-term change in riverine Hg export to be possible from the current river monitoring programme, an analytical method with a lower detection limit would be necessary.

The role of aquatic processing for retention of Hg from headwaters to the coast is difficult to assess with the values for catchment export produced in this report. The summed aquatic loading (headwater export and atmospheric deposition) is higher (546 kg) than the estimated Hg inputs to the coast (281-331 kg), which points to significant retention and/or losses of Hg on its way from headwaters to coast. Retention of Hg along the aquatic continuum is difficult to quantify, mainly because headwater Hg inputs to surface waters are very uncertain at the regional scale. For small catchments, Hg fluxes can be estimated with more confidence as the data from ICP IM demonstrate. Thus, to evaluate impacts of changes in atmospheric emissions of Hg on aquatic ecosystems, it is important to distinguish between catchment and atmospheric loads of Hg to surface waters. Catchment loads are a result of current Hg and legacy Hg deposition, and are likely to respond more slowly to lower atmospheric concentrations than direct inputs to lakes through wet deposition.

4 Conclusion

Nearly two centuries of elevated Hg deposition from anthropogenic activities have led to considerable stores of Hg in catchment soils and high concentrations in aquatic food webs, even in remote locations. In Norway, concern relates primarily to elevated concentrations of Hg in freshwater fish, many places exceeding maximum limits for protection of human health. A future reduction in atmospheric loading of Hg may lead to reductions in aquatic concentrations and riverine transport of Hg to coastal areas, but sparse data limit assessments of how changes in atmospheric Hg impact aquatic ecosystems.

A Hg budget is calculated for Norway by estimating fluxes of Hg in the environment. Atmospheric deposition (wet and dry) for forest, open land, and water, and riverine export of Hg are estimated using various methodological approaches. Atmospheric inputs (3077 kg) are a factor 10 higher than riverine export (281-331 kg), indicating significant retention of Hg likely occurring in soils and in surface waters. To estimate environmental loading of Hg to surface waters, we distinguish between headwater (=catchment) export of Hg and atmospheric Hg loadings. By linking headwater organic carbon export to Hg-to-carbon ratios from lake and river monitoring, a total mass flux of 295 kg Hg from headwaters was estimated. This estimated headwater export is similar to the riverine export of Hg (281-331 kg), suggesting that retention of Hg along the aquatic continuum is of similar size as the direct atmospheric inputs to surface waters (251 kg). The similarity of the headwater export and surface waters deposition estimates could imply that reductions in wet deposition of Hg would have positive implications for Hg accumulation in the aquatic foodweb. The headwater Hg export is however uncertain at the regional scale, and Hg budgets for small headwater catchments are more reliable and valuable to assess the fate of atmospheric Hg, and the role of aquatic Hg retention.

Given the limited understanding of Hg cycling and availability of monitoring data for deposition, catchment export, and retention of Hg, we recommend continuation of catchment and river monitoring of Hg, and stimulation of international collaboration on Hg monitoring and modelling. The quantification of key elements in the Hg budget for Norway is useful for highlighting available data sources and data gaps, and we conclude that Hg budgets at national, regional, and local scales are valuable tools for impacts assessments of Hg in the environment.

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