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Modelled Nitrosamine and Nitramine concentrations in Lake Elvåga following amine-based CO_2 Capture at FOV Waste Incineration Plant at Klemetsrud

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Modelled Nitrosamine and Nitramine concentrations in Lake Elvåga following amine-based CO_2 Capture at FOV Waste Incineration Plant at Klemetsrud

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1 NIVA MODELLED NITROSAMINE AND NITRAMINE CONCENTRATIONS IN LAKE ELVÅGA FOLLOWING AMINE-BASED CO2 CAPTURE AT FOV WASTE INCINERATION PLANT AT KLEMETSRUD

NE/CERC have performed dispersion and deposition modelling of NO_2, nitrosamines and nitramines from the planned CC-plant at Klemetsrud (NC03i-FOV-S-RA-001). As a part of the work rough calculations of drinking water concentration of nitrosamines (NSAs)and nitramines (NAs) was done, but it was recommended to do more detailed catchment modelling.

NIVA has been engaged to do catchment modelling. The advanced catchment and lake water modelling simulated lake water concentrations of NSAs and NAs in Lake Elvåga following potential amine-based CC at the nearby waste incineration plant at Klemetsrud and are based on dry and wet deposition calculated by NE/CERC.. The combined catchment and lake model produced a good fit for the site-specific hydrological and climatic variables which indicates a high degree of site specificity of the simulations.

Catchment and lake processes were found to have a large impact on NSA and NA lake water concentrations and to be highly impacted by seasonal climatic events. While the deposition rates of NSAs were higher than for the NAs, final lake water concentrations after the three-year modelled period showed the opposite picture. Efficient photodegradation of the NSAs was the explanation, almost balancing out the input rates. With the absence of an efficient depletion pathway for the NAs, levels were found to accumulate over time, both in the hypolimnion and epilimnion of the lake.

Seasonal variation had a strong impact on the photodegradation rates of NSAs and on the input rates of both NSAs and NAs. During winter the effect of photodegradation was reduced to a minimum, resulting from the combined effect of weaker sunlight radiation and the presence of an ice cover on the lake. Accordingly, winter-concentrations of NSA were more than five times higher than during the rest of the year. Input rates of both NSAs and NAs were highest during spring, resulting from increased transport to the lake with melting snow. For the NSAs, however, this effect was completely cancelled out by high photodegradation rates during spring.

For the three year period modelled (2015-2017), maximum annual average concentrations of NSAs and NAs amounted to 0.24, 3.3, and 4.4 ng L-1 for the “best”, “likely”, and “worst case scenarios, respectively. These values represent the range of uncertainty in the model result, with values toward the two edges of the interval being less likely than value in the middle. The uncertainty associated with the biodegradation rate was found to be the most important cause of uncertainty in the lake concentrations. The biodegradation rate used in the likely scenario was the one most commonly reported in the literature, while the range used for the other scenarios represent other reported values.

Continued deposition over 10 years seemed to stabilise the annual average value to around 3.5 ng L-1, in the likely case. The high impact from seasonal variation should be taken into consideration for the planning of potential future monitoring programmes. Moreover, a problem related to future monitoring is the lack of sufficiently sensitive methods for the determination of NSAs and NAs in natural waters. In particular, for the NAs, there is no method available to measure concentrations around 4 ng L-1 or below. To further reduce the uncertainty of the modelled estimates, future work should focus on assessing the biodegradability of the NAs

since this was the parameter found to have the largest impact on the lake NSA and NA concentrations.

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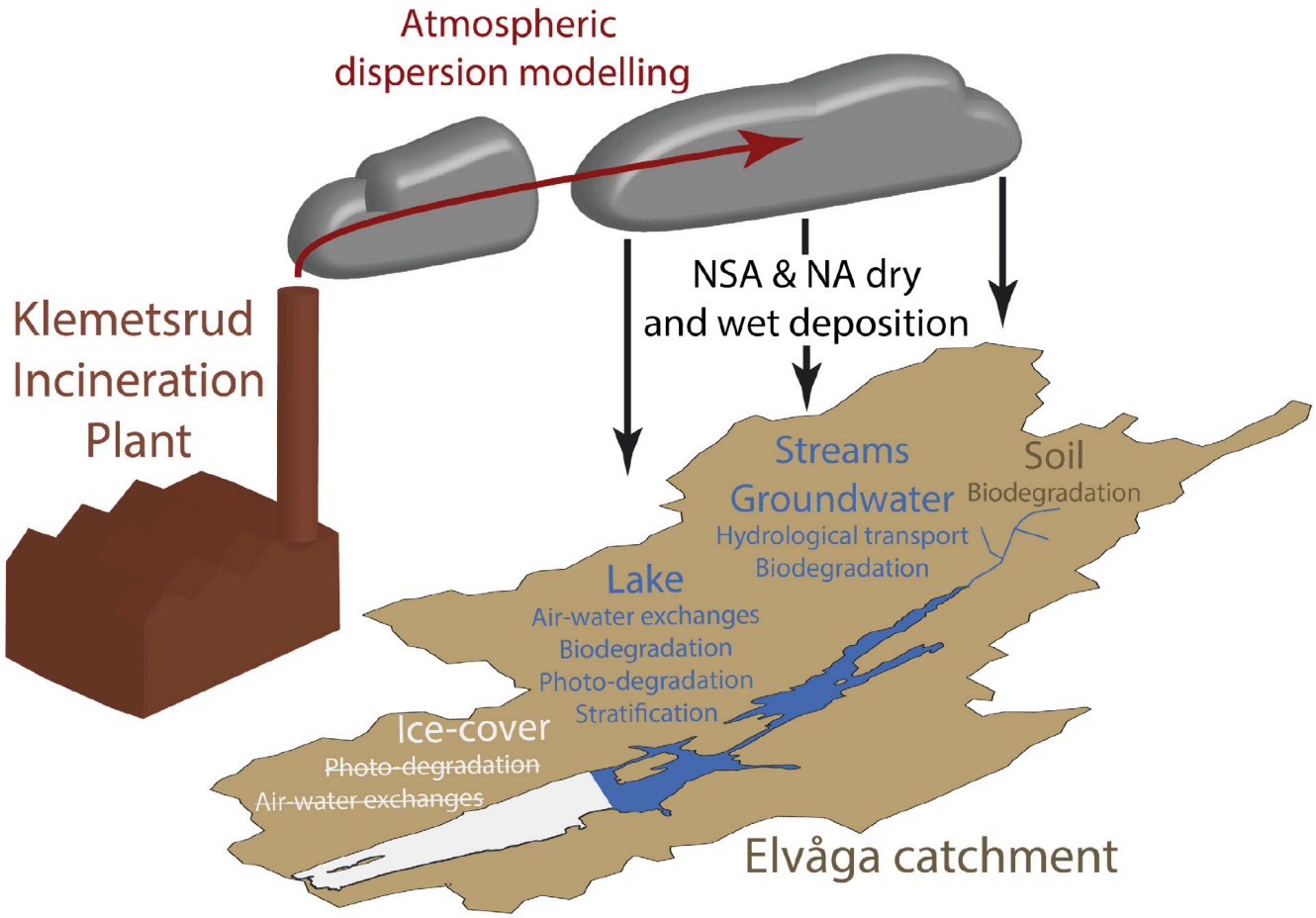
Modeled Nitrosamine and Nitramine concentrations in Lake Elvåga following amine-based CO_2 Capture at FOV Waste Incineration Plant at Klemetsrud



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Summary

Future levels of the carcinogenic- and potentially carcinogenic nitrosamines (NSAs) and nitramines (NAs), respectively, were modelled in the drinking water source, Lake Elvåga following the planned full-scale amine-based CO_2 capture at the Oslo waste incineration plant. A high-resolution and dynamic catchment model (INCA-Contaminants) was combined with a lake model add-on. NSA and NA deposition rates (provided from COWI/Norsk Energi/CERC) were combined with site specific information (hydrology and climate), and literature values of NSA and NA bio- and photodegradation rates and physiochemical parameters. The three scenarios, “best”, “likely”, and “worst” cases were created to capture the uncertainty of key parameters. Three years of full-scale CO_2 capture resulted in maximum annual average sums of NSAs and NAs in the lake water at 0.24, 3.3, and 4.4 ng L^{-1}, for the three scenarios, respectively. For the NSAs, photodegradation was an efficient depletion pathway, nearly balancing out the input rates. Lake water levels of NAs increased with time due to the lack of an efficient depletion pathway. Seasonal variation was evident in both the NSA and NA concentrations and this should be considered for potential future monitoring programs.

Four keywords	Fire emneord
1. Catchement model	1. Nedbørfelt modell
2. CO_2 capture	2. CO_2 fangst
3. Nitrosamines	3. Nitrosaminer
4. Nitramines	4. Nitraminer

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Modelled Nitrosamine and Nitramine concentrations in Lake Elvåga following amine-based CO_2 Capture at FOV Waste Incineration Plant at Klemetsrud

Preface

The catchment and lake modelling to estimate annual concentrations of nitrosamines (NSAs) and nitramines (NAs) in Lake Elvåga was conducted by the Norwegian Institute for Water Research (NIVA) on commission from COWI. Tore Methlie Hagen was COWI's contact person for the project. Dag Borgnes from Norsk Energi provided the data from the atmospheric dispersion and deposition modelling. Claus Jørgen Nielsen from the Department of Chemistry, University of Oslo contributed with physiochemical descriptors of the NSAs and NAs.

The catchment modelling was conducted by Magnus D. Norling, the lake modelling by François Clayer, and with contribution from Jose-Luis Guerrero. Cathrine Brecke Gundersen was the project manager and contributed with experiences in the catchment processing of NSAs and NAs.

Yan Lin was responsible for the quality assurance of the work and the project.

Oslo, 09.06 2020

Cathrine Brecke Gundersen

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Summary

According to the international panel on climate change, implementation of CO_2 capture technology is needed to reach the climate goal of limiting global warming to 2 °C by 2100. In Norway, the major CO_2 point-source emitters, including the waste incineration plant at Klemetsrud in Oslo (Fortum Oslo Varme), are currently under evaluation for their suitability for CO_2 capture. The currently most feasible way of capturing CO_2 is using amines which can lead to the formation of the carcinogenic- and potentially carcinogenic nitrosamines (NSAs) and nitramines (NAs). Drinking water sources are of particular concern since the NSAs and NAs have a strong affinity for the aqueous phase. Accordingly, the Norwegian Institute for Public Health has issued a safety limit for the sum of NSAs and NAs in drinking water at 4 ng L^{-1}. Previously conducted atmospheric modelling found that the Lake Elvåga, a nearby drinking water source, was susceptible to receive NSAs and NAs from the planned CO_2 capture at the waste incineration plant in Oslo.

The aim of this study was to simulate levels of NSAs and NAs in Lake Elvåga by assessing the effect from various hydrological, climatic, and biogeochemical parameters. This was achieved by linking a dynamic and high-resolution catchment model (INCA-Contaminants) in combination with a lake-model add on (developed in-house). NSA and NA deposition rates (provided from COWI/Norsk Energi/CERC) were combined with site specific information (hydrology and climate), and literature values of NSA and NA bio- and photodegradation rates and physiochemical properties. Three different scenarios were created to capture the uncertainties related to some of the key parameters, and constituted “best”, “likely”, and “worst” cases. A period of three years (2015 – 2017) was modelled to investigate both the potential for accumulation with time and the influence from seasonal climatic variation.

Three years of full-scale CO_2 capture reached maximum annual average sums of NSAs and NAs in the lake water at 0.24, 3.3, and 4.4 ng L^{-1} for the “best”, “likely”, and “worst” cases, respectively. For the three scenarios, the parameters found to be associated with some uncertainty at the same time as having a large impact on the lake water concentrations, were selected to be varied. Thus, the “best”- and “worst”-case scenarios then represent the lowest and highest annual mean concentrations that can be expected based on the current knowledge. It should be noted these two scenarios can be considered unlikely as they represent situations where all parameter values are simultaneously at their extremes. The likely case represents values toward the centre of the available parameter ranges. For parameters where there were no data available, conservative estimates were made based on the precautionary principle. The main cause for the lower level in the “best” case scenario was a higher biodegradation rate for the NAs. It is important to be aware that the biodegradation rates have very high uncertainties due to the lack of sufficient studies available in the literature, and the lake concentration is also highly sensitive to this value. For the NSAs, photodegradation was an efficient depletion pathway, nearly balancing out the input rates. In contrast, NAs levels in the lake were found to accumulate with time, due to the lack of an efficient depletion pathway.

Seasonal variation in temperature, sunlight, and hydrology was found to influence both the NSA and NA concentrations and this should be acknowledged in any potential future monitoring programs, in addition to the need for developing sufficiently sensitive analytical methodology. To further reduce the uncertainty of the estimates, future studies should focus on establishing the site-specific biodegradation rates of NAs.

Sammendrag

Tittel: Modellerte nitrosamin og nitramin konsentrasjoner i innsjøen Elvåga i sammenheng med amin-basert CO_2 fangst på FOV forbrenningsanlegg på Klemetsrud
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Forfatter(e): Cathrine Brecke Gundersen, Magnus Dahler Norling og François Clayer
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Ifølge FNs klimapanel (IPCC) vil teknologi for CO_2 fangst være nødvendig for å nå klimamålet om å forhindre global oppvarming til en økning på 2 °C innen 2100. I Norge vurderes nå de største punktkildene til CO_2-utslipp, inkludert forbrenningsanlegget på Klemetsrud i Oslo, for mulighet til å etablere CO_2 fangst. Metoden som benyttes til å fange CO_2 baserer seg på aminer som kan føre til dannelse av henholdsvis kreftfremkallende- og mulig kreftfremkallende nitrosaminer (NSAs) og nitraminer (NAs). Det norske Folkehelseinstituttet (NIPH) har kommet med en anbefalt drikkevannsgrense på 4 ng L^{-1} for summen av NSAs og NAs. Ettersom NSAs og NAs er svært løselige i vann vil utslipp i nærheten av drikkevann kunne utgjøre en risiko for folkehelsen. Tidligere utført atmosfærisk modellering har vurdert avsetning til ulike nærliggende drikkevannskilder. Innsjøen Elvåga ble da funnet å være mest utsatt for å motta NSAs og NAs fra Klemetsrud dersom CO_2 fangst blir iverksatt.

Hensikten med dette studiet var å estimere konsentrasjoner av NSAs og NAs i Elvåga ved å undersøke mulig påvirkning fra lokal hydrologi, klima og biogeokjemiske prosesser. Dette ble gjort ved å kombinere en dynamisk og høyoppløselig nedbørfeltmodell (INCA-Contaminants) med en innsjømodell (utviklet av NIVA). Nedfallsrater av NSAs og NAs (gjort tilgjengelig av COWI/Norsk Energi/CERC) ble kombinert med stedsspesifikk informasjon om hydrologi og klima, og litteraturverdier som beskriver NSA og NA bio- og fotonedbrytning samt deres fysiokjemiske egenskaper. Tre ulike scenarioer ble laget for å dekke usikkerheten som var knyttet til enkelte av parameterne. Scenariene utgjorde «best», «sannsynlig» og «verste fall». Modellen ble kjørt over en periode på tre år (2015 – 2017) for å kunne undersøke mulig akkumulering over tid samt mulig påvirkning fra sesongvariasjoner.

Etter tre år med modellert fullskala CO_2 fangst på Klemetsrud var maksimum årlige gjennomsnitt for summen av NSAs og NAs i Elvåga på 0,24, 3,3 og 4,4 ng L^{-1} for de tre ulike scenariene “best”, “sannsynlig” og “verste fall”. Som bakgrunn for de tre scenariene ble parametere valgt ut til å varieres dersom de var knyttet til usikkerhet og samtidig hadde en stor innvirkning på sluttkonsentrasjon. Scenariene «best» og «verste fall» utgjorde dermed det laveste og høyeste årlige gjennomsnittet som kunne forventes basert på gjeldende kunnskap. Merk at disse to scenariene anses som usannsynlige ettersom de representerer situasjoner der alle parameter verdiene samtidig er ved deres ytterpunkter. «Sannsynlig»-scenariet representerer parameter verdier som er i midten av deres tilgjengelige områder. For de parameterne der det ikke var data tilgjengelig ble en konservativ verdi valgt, basert på best tilgjengelig kunnskap. Hovedårsaken til den lavere sluttkonsentrasjonen i «best» utfall scenariet var en høyere bionedbrytningshastighet for NAs. Denne verdien var meget usikker på grunn av de få studiene som tidligere har blitt utført for å undersøke NA bionedbrytning. Samtidig ble NA bionedbrytning funnet til å ha en stor innvirkning på konsentrasjonene i innsjøen. For NSAs var fotonedbrytning en effektiv nedbrytningsprosess som nærmest fjernet all innkommende NSAs. Ettersom det ikke foreligger noen dokumentert effektiv nedbrytningsprosess for NAs vil disse akkumulere i innsjøen over tid.

Sesongvariasjoner i temperatur, sollys og hydrologi påvirket nivåer av både NSA og NA i innsjøen, og dette bør vurderes i planleggingen av fremtidig overvåkningsprogram, i tillegg til nødvendigheten av å utvikle følsom analytisk metodikk. For å kunne redusere usikkerheten knyttet til estimatene anbefales det at fremtidige studier bør fokusere på bionedbrytning av NAs.

1 Introduction

Technology for CO_2 capture (CC) is a promising mitigation tool that must be implemented to reach the climate goals of limiting global warming to 2 °C by 2100 (IPCC, 2014). The principle is to isolate CO_2 out from the flue gas of large scale point emitters (> 0.1 Mt CO_2 $year^{-1}$) like oil refineries, cement factories, or incineration plants and to subsequently either store the concentrated CO_2 safely underground or to reuse it in some industrial process (IPCC, 2005). Currently, the most successful method of capturing CO_2 is by using amines (Rochelle, 2009). However, concern is raised since small amounts of amines can escape the CC plant with the cleaned flue gas. In the atmosphere, the amines will rapidly degrade through sunlight driven reactions to form the carcinogenic- and potentially carcinogenic nitrosamines (NSAs) and nitramines (NAs), respectively (Mazari et al., 2019; Reynolds et al., 2012). The Norwegian Institute for Public Health (NIPH) has recommended a guideline limit of 4 ng L^{-1} for the sum of NSAs and NAs in drinking water (and 0.3 ng m^{-3} in air) (Låg et al., 2011). While the presence of the precursor molecule, amines, have been documented at low levels in natural waters, measurements of relevant NSAs and NAs remains to be made (Poste et al., 2014). One major limitation to the study of NSAs and NAs in natural waters is the limited analytical methods available for their determination. For the NAs, in particular, there exist no method capable of measuring at the NIPH level (Gundersen, Cathrine Brecke et al., 2017).

The waste incineration plant in Oslo (Fortum Oslo Varme) is the largest CO_2 point emitter in Oslo. The site is currently under evaluation and testing for its suitability to instalment of CC technology. The waste incineration plant is located close to densely populated areas of the city of Oslo, including lakes that serves as drinking water sources (Lakes Elvåga (10 %), Maridalsvannet (90 %), and with Alnsjøen and Langlia as reserve sources). Atmospheric dispersion and deposition modelling has previously been conducted by Norsk Energi and Cambridge Environmental Research Consultants, using different emission scenarios and weather conditions, to predict future atmospheric concentrations and deposition rates of NSAs and NAs at the nearby area. Using settings representative for a worst-case scenario, levels of NSAs and NAs were found to exceed the NIPH threshold in the nearest lake, Lake Elvåga (Norsk Energi, 2019). However, the estimated lake water concentration was made on very simplified assumptions regarding catchment and lake hydrology as well as potential biogeochemical processing.

Once the NSAs and NAs are deposited on ground, several processes will interplay to determine final lake water concentrations. The physiochemical properties of the molecules (Henry's law constant, octanol-water partitioning coefficient, etc.) will govern their partitioning between air, soil, and water. Both the NSAs and NAs are very hydrophilic and are thus expected to mainly reside in the aqueous phase. Accordingly, low soil sorption potential has been found for both compound groups (Gundersen, C. B. et al., 2017; Gunnison et al., 2000). Overall, the NSAs are of higher volatility than the NAs, and some NSAs are sufficiently volatile to escape from the water phase to air. NSAs and NAs dissolved in water will be transported under various hydrological flow regimes in the catchment, e.g. with overland flow, soil water runoff, and groundwater into a branched river network before entering the lake. Along this pathway, and once in the lake, the NSAs and NAs will be subject to a number of biogeochemical processes. Photodegradation is an important depletion pathway for the NSAs, both in air and in water, while the NAs are not prone to photodegradation (Sørensen et al., 2015). Biodegradability rates are expectedly low for both compound classes (Brakstad et al., 2018). Seasonal variation in e.g. precipitation and sunlight will further influence these processes.

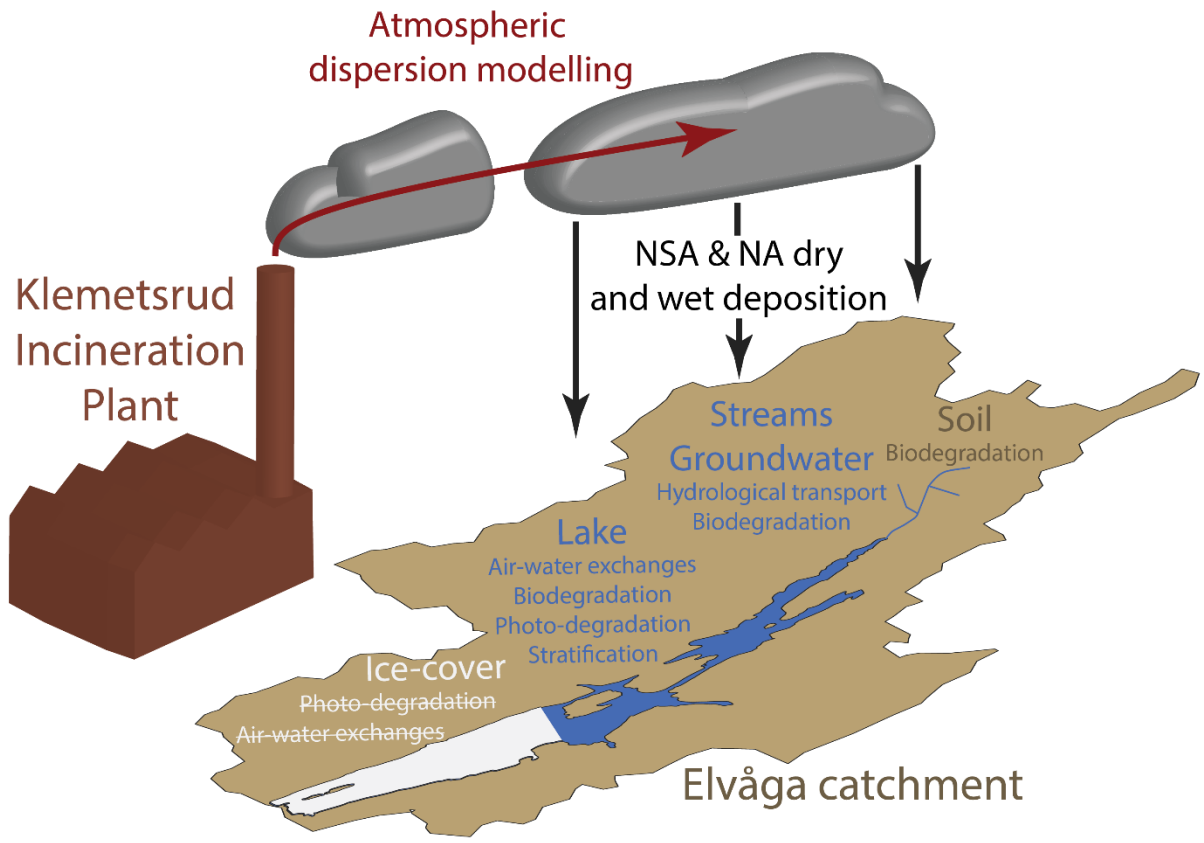

Figure 1: Schematic representation of key processes influencing lake water concentration of NSAs and NAs including 1) atmospheric emission, transport, and deposition (previously modelled by Norsk Energi / Cambridge Environmental Research Consultants) and 2) catchment processes covering hydrological transport, air-water exchange, biodegradation and photodegradation, in addition to the effect from seasonal events such as lake ice-cover.

The objective of this study was to estimate annual lake water concentrations of NSAs and NAs in Lake Elvåga following full-scale amine-based CC at the waste incineration plant in Oslo. Atmospheric concentration and deposition rates, provided by Norsk Energi (2019), served as the input. The catchment processes illustrated in Figure 1 were considered for their influence on final NSA and NA lake water concentration. This was accomplished using the INCA-Contaminants catchment model (Nizzetto et.al. 2016) in combination with a lake model add-on (developed in-house). This enabled the site-specific simulation of NSAs and NAs movement through the catchment, including phase transfer and biogeochemical processing, to estimate final lake water concentrations. Three different scenarios were included to constitute “best”, “likely”, and “worst” cases (ref. 2.4) to capture the uncertainty in key parameter values. The model was run for three consecutive years (2015 – 2017) to study potential accumulation with time.

2 Methods

2.1 Input Data

2.1.1 Atmospheric Concentrations and Deposition Rates

Data of NSA and NA atmospheric concentrations and deposition rates, estimated from previously conducted atmospheric dispersion and deposition modelling, was provided by Norsk Energi (Norsk Energi, 2019). The data represented their "Reheat" case scenario (representing future operating situation) and was of hourly resolution for the year 2016. The NSAs and NAs were treated as two generic molecular groups. The data was aggregated to daily averages which was the resolution set for the catchment/lake model. See Table 1 for the sum of dry and wet deposition of nitrosamines and nitramines.

Table 1: Average values of the daily sum of dry and wet deposition of nitrosamines and nitramines. Data provided from Norsk Energi (2019).

Parameter	Unit	Nitrosamines (NSAs)	Nitramines (NAs)
Sum dry + wet deposition	$ng/m^2/day$	4.139	8.241

2.1.2 Molecular Physiochemical Properties

In accordance with the atmospheric modelling work, the NSAs and NAs were treated as two distinct compound groups. The values set for the physicochemical parameters are listed in Table 2 and constitutes the averages of the most studied NSAs and NAs (N-nitrosodimethylamine, dimethylnitramine, monomethylnitramine, dimethylnitramine, and monoetanolnitramine) as well as highly relevant but censored NSAs and NAs, provided by prof. emer. Claus J. Nielsen. The enthalpy of phase transfer between air and water was the only parameter not known, and was therefore set to vary within a range that is typical for semi-volatile substances (Schenker et al., 2005; Shen & Wania, 2005).

Table 2: Selected parameters for physical and chemical properties of nitramines and nitrosamines.

Parameter name	Abbr.	Unit	Nitrosamine	Nitramine
Molar mass*	Mm	$g\ mol^{-1}$	74.1	90.7
Molecular volume**	Mv	$cm^3\ mol^{-1}$	56.5	90.7
Henry's constant at 25°C*	H	$Pa\ m^3\ mol^{-1}$	0.104	0.081
Octanol-water partitioning coefficient*	$\log_{10} K_{OW}$		-1.7	-0.6
Enthalpy of phase transfer between air and water***	ΔH	$kJ\ mol^{-1}$	5-50	5-50

*Calculated using EPISUITE software.

**Assuming a density of 1 and 1.31 for the NAs and NSAs, respectively (prof. emer. Claus J. Nielsen).

***Typical values for semi-volatile substances (Schenker et al., 2005; Shen & Wania, 2005).

2.1.3 Climate

Historical climatic data from the years 2015 to 2017 was used in the modelling work, in accordance with the previously conducted atmospheric modelling. Precipitation, air temperature, solar radiation, and wind speed originated from the Blindern meteorological station while solar radiation came from the Arnes station. Air pressure and relative humidity was provided from ERA5 reanalysis data (Copernicus Climate Change Service Climate Data Store).

2.2 Numerical Models

A catchment model was combined with a lake model to simulate the concentrations of NSAs and NAs deposited both on catchment soil and lake water, including the effect from the various biogeochemical processes in both compartments. The study was conducted at daily time steps for three consecutive years to assess the potential accumulation of the compounds with time. This moreover enabled a seasonal assessment of NSA and NA in lake water. The climatic data spanned the three years from 2015 till 2017, while the atmospheric concentration and deposition data from 2016 was repeated for this three-year period. Due to the lack of measured concentrations to compare modeled results with, it was not possible to do any formal uncertainty analysis. Instead, the values of the most sensitive parameters were varied within ranges reported in the literature to estimate the uncertainties (see section 2.4).

The model itself (but not any of the data used for this report) is open source as part of the Mobius framework, and the specific implementation can be viewed at:
https://github.com/NIVANorge/Mobius/blob/master/Modules/INCA-Tox.h
https://github.com/NIVANorge/Mobius/blob/master/Modules/INCA-Tox-Lake.h

2.2.1 Catchment Model

The INCA-Contaminants model (Nizzetto et al., 2016) is a high-resolution and dynamic catchment model that builds on the hydrology model PERSiST (Futter et al., 2014). In general, contaminants enter the soil through atmospheric deposition, are dynamically partitioned between solid and aqueous phases in the soil, groundwater and river, and transported along the various water flow paths. Degradation in all phases and compartments is computed. The model is fed with site-specific numeric information, including catchment characteristics and climatic conditions to establish a site-specific scenario. By providing data on contaminant deposition rates, the final concentration of the contaminant at a selected place in the catchment (here: lake) can be estimated.

2.2.2 Lake Model

The sum of the NSAs and NAs transported from the catchment to the lake and deposited directly onto the lake served as input to the lake model. The model considers influence from site specific factors such as lake morphology, degradation processes (photodegradation and biodegradation), and seasonal climatic variation. Water temperature can have a large impact on final lake water NSA and NA concentration in the sense of thermal stratification and the formation of an ice cover. In the model, we considered two layers: i) the lower section of the lake, i.e., the hypolimnion, with a constant temperature of 4.5 °C, and ii) the top 5 meter of the water column, i.e., the epilimnion, where water temperature was computed using inflow water temperature and the net heat flux from radiation, latent and sensible heat. Standard attenuation of shortwave radiation due to water turbidity was computed using measured secchi depth (simplified from Kondo, 1975). The net heat flux model was based on the one in the GOTM model (Burchard et al., 1999; Kondo, 1975). When epilimnion and hypolimnion temperatures were less than 1°C apart the water masses of both layers were mixed, mimicking seasonal lake turnover (spring and fall). All NSAs and NAs entering the lake from the catchment were assumed to be completely dissolved into the epilimnion. The formation of an ice-cover during winter was also considered since it impacts photodegradation through attenuation of short-wave radiation as well as diffusive exchange (Henry's law) of compounds between the lake surface and the atmosphere. This modelling procedure provided acceptable estimations of the temperature compared to the measured values reported by Isidorova et al. (2016).

2.3 Parametrisation

2.3.1 Catchment Hydrology

The soil and groundwater hydrology of the model (PERSiST) was calibrated using existing data from the nearby catchment, Mariholtputten, for which runoff data was available from The Norwegian Water Resources and Energy Directorate (NVE) for the period from 1997 to 2002. This was done since there was no suitable daily runoff data available for the Elvåga catchment. Selected hydrological parameters are presented in Table 3.

Table 3: Selected hydrological parameters for the catchment model. Note that for the Degree-day evapotranspiration and Baseflow index, a range of different values were used to capture the uncertainty of the parameters.

Parameter name	Value	Unit
Degree-day evapotranspiration*	0.09 - 0.20	mm/°C/day
Soil water time constant	5.7	Days
Groundwater time constant	75	Days
Baseflow index*	0.1 - 0.6	
Catchment area	17.07	km^2

** Due to the high uncertainty and high impact associated with these parameters, they were selected to be varied in the three scenarios.*

The degree-day evapotranspiration has a direct impact on soil NSA and NA concentration. By removing soil moisture, the contaminant will be concentrated in the residual soil water. Hence, higher evapotranspiration tends to increase lake contaminant concentration. The value of 0.14 gave the best fit for predicting runoff in the Mariholtputten case, which is close to typical values frequently used for similar catchments (unpublished data). However, some uncertainty was associated with this value since some discrepancies between rainfall and observed flow in the catchment was observed, and both rainfall and evapotranspiration had to be scaled to obtain a satisfactory fit to the data. The value of 0.09 was needed to simulate an average 2015-2017 runoff close to the 1961-1990 average runoff of 0.35 m^3s^{-1} reported by NVE (NVE, 2006). It is reasonable that runoff could be lower in 2015-2017 comparted with 1961-1990 due to higher temperatures, and low rainfall in 2016. There could also be discrepancies between Blindern measurements and the actual precipitation in the catchment. The degree-day evapotranspiration parameter was varied in the scenarios (see below) because of the above-mentioned uncertainties and because compound concentrations in water were sensitive to it. **The soil and groundwater time constants** determine how fast water flows through the soil and groundwater, respectively. These were calibrated based on the Mariholtputten case. **The baseflow index** controls the amount of the soil water running through the groundwater compartment before entering the lake. This will have an impact on the lake water concentration since the water residence time in the groundwater is much larger than in the surface waters (e.g., soil and river; see Table 3), thereby providing more time for biodegradation. Hence, a higher baseflow index will usually reduce the concentration of contaminants in the lake. **The catchment area** was the total surface area of land where precipitation drains into Elvåga.

2.3.2 Lake Hydrology

Selected parameters for the lake model are presented in Table 4. The lake was modelled as having a conical volume, with the upper 5 meters belonging to the epilimnion and the rest to the hypolimnion (maximum 46.3 m). The lake water volume was considered constant since the lake is regulated by the water works of Oslo municipality and the water table rarely varies by more than one meter (Water and wastewater, Oslo municipality, Pers. Comm. 2020).

Table 4: Selected parameters of the physical lake model.

Parameter name	Value	Unit	Reference
Surface area	1.67	km^2	
Max depth	51.3	m	Isidorova et al. (2016)
Epilimnion thickness	5	m	
Secchi depth	2.5	m	

2.3.3 Aqueous Phase Removal Processes

The **NSAs photodegradation rate** in the lake epilimnion ($R_{photo}^{Nitrosamine}$; ng day^{-1}) was formulated as:

$$R_{photo}^{Nitrosamine} = -OC_{Nitro} \times QY_{Nitro} \times \frac{f_{UV}}{e_{UV}} \times 86400 \times NSA \times Q_{sw} \times (1 - Albedo) \times Attn$$

where NSA is the total mass of NSAs in the epilimnion (ng), OC_{Nitro} is the average optical cross-section of nitrosamine around 340 nm (m^2 mol Nitro $^{-1}$), QY_{Nitro} is the quantum yield describing the number of NSA molecule transformed by number or photon absorbed (mol Nitro mol $photon^{-1}$), f_{UV} is the fraction of UV radiation in sunlight (unitless), e_{UV} is the energy of a UV photon at 340 nm (J mol $photon^{-1}$), Q_{sw} is the incoming short wave radiation at the lake surface (W m^{-2}), $Albedo$ is the water or ice albedo (unitless), and $Attn$ is the light attenuation coefficient in the water column calculated from Secchi depth (see Table 5). NSA absorption spectra show two peaks at about 230 nm and 340 nm (Afzal et al., 2016), of which only the second is within the sun light spectrum at sea level (Moan, 2001). The absorbance reported at 340 nm is typically 100 times lower than at 230nm (Afzal et al., 2016; Shim et al., 2016). The effect of winter ice-cover on photodegradation was considered by shadowing incoming radiation during the relevant time period through the $Albedo$.

The **biodegradation rate** (R_{bio}; ng day^{-1}) was formulated as follows for both NSAs and NAs, in both the catchment and lake compartments:

$$R_{bio} = Compound \times k_{bio} \times \theta^{(T_{water}-20)}$$

Where $Compound$ is the NSA and/or NA mass in a given compartment (ng), k_{bio} is the biodegradation constant at 20°C (day^{-1}), θ is the temperature adjustement coefficient (unitless), and T_{water} is the water temperature (°C) in the given compartment. The effect of temperature on biodegradation was considered through the parameter θ (standard value of 1.05) which scales the biodegradation by 5% for each temperature change of 1°C, i.e., at 21°C the biodegradation rate will be 5% larger than at 20°C. The same value of $k_{bio}$was used for all compartments since it has been shown that aerobic and anaerobic biodegradation rates are similar (Brakstad et al., 2018).

Potential influence from **NSA and NA sorption** to lake sediments was not included since the necessary catchment-specific information (e.g. bed sediment mass) was not available. For the same reason, transport via soil erosion from the catchment was turned off in the model. Given the high solubility of NSAs and NAs in water, and the low soil sorption rates found in the literature (Gundersen, C. B. et al., 2017; Gunnison et al., 2000), loss of NSAs and NAs from the aqueous phase due to sorption to solids should represent a negligible fraction. Another potential depletion pathway of soil NSAs and NAs that was not considered is plant uptake (Panz & Miksch, 2014). Given the high

nitrogen content in these compounds, plants could potentially remove some from the soil before being transportation with runoff to the lake. This was not included due to the complete lack of data for the compounds in question.

Table 5: Selected parameters of the biogeochemical model.

Parameter name	Value	Unit	Reference
k_{bio} (at 20°C)*	0 - 0.05	day^{-1}	Brakstad et al. (2018)
θ	1.05	-	Bowie et al. (2004)
OC_{Nitro}*	1.2 - 5.0	m^2 mol Nitro^{-1}	Shim et al. (2016)
QY_{Nitro}	0.45	mol Nitro mol photon^{-1}	Sørensen et al. (2015)
f_{UV}	0.06	-	Saloranta and Andersen (2007)
e_{UV}	351843	J mol photon^{-1}	
$Albedo$	0.04 (free water) 0.6 (ice covered)	-	
$Attn$	0.29	-	Calculated from Secchi depth

** Due to the high uncertainty and high impact associated with these parameters, they were selected to be varied in the three scenarios.*

2.4 Possible Scenarios

Due to uncertainties in some parameter values, we chose to run the model for three different scenarios, considered "best", "likely", and "worst" cases in order to cover possible outcomes of different parameter combinations. The parameters chosen to be varied are presented in Table 6 and were selected both for having a high uncertainty and for the uncertainty having a significant impact on the lake NSA and NA concentrations (sensitivity). The three different values were set based on literature values and own experiences with modelling similar type of catchments, discussed in Section 2.3.1 and 2.3.2. Thus, the three scenarios are intended to capture the uncertainty in lake concentrations caused by uncertainties in parameter values. The sensitivity, also presented for each parameter in Table 6, indicates how much the average lake concentration (NSAs + NAs) would increase if the value of that specific parameter was changed from the "best" to the "worst" case-value while keeping all other parameter values at the "likely case".

Table 6: Selected parameter values for "best", "likely", and "worst" case scenarios. The sensitivity (%) indicates how much each parameter would influence the 2017 mean lake concentration (NA+NSA) increases when moving the value of a parameter from the "best" value to the "worst" value while holding other parameters constant at the "likely" value.

Parameter name	Unit	Best	Likely	Worst	Sensitivity
Baseflow index	-	0.6	0.2	0.1	1%
Degree-day evapotranspiration	mm/°C/day	0.09	0.14	0.20	20%
k_{bio} (at 20°C)	day^{-1}	0.05	0.001	0.000	1068%
OC_{Nitro}	m^2 mol Nitro^{-1}	5	2.5	1.2	4%
Diffusive exchange multiplier	-	1	1	0.5	15%
Enthalpy of phase transfer between air and water	kJ mol^{-1}	5	50	50	4%

The parameters selected covered catchment hydrology, aqueous removal processes, in addition to two parameters related to the diffusion of NSAs and NAs between the water phase and air. Here follow the considerations that were made when setting the parameter values for the three scenarios. See above for explanations of the different parameters.

With the absence of data on surface discharge into the Lake Elvåga, **the baseflow index** could not be constrained. Instead, the values of 0.6 was selected for the "best", 0.2 for the "likely" and 0.1 for the "worst" case scenarios, representing typical minimum and maximum values for this type of catchment (unpublished data). Through the parametrization of the hydrological model, several different values for the **Degree-day evapotranspiration** were found to be plausible**.** The values of 0.09, 0.14, and 0.20 were selected for the "best", "likely", and "worst" case scenarios, respectively. These values were selected in accordance with the daily runoff data from a nearby catchment over the period 1997-2002 and with the average yearly runoff of 0.35 m^3/s reported by NVE over 1961-1990 (NVE, 2006) (see earlier discussion). In the literature, different values have been reported for the NSA and NA **biodegradation rate constants (k_{bio})**, ranging from < 0.001 to 0.09 day^{-1} (Brakstad et al., 2018, and references therein). Most frequently low biodegradation rates (< 0.001) have been observed, and thus, the value of 0.001 was selected for the "likely" case. For the "best" and the "worst" case scenarios, values of 0.05 and 0 were set, respectively. These values reflect ranges of reported values for both NAs and NSAs. **UV-light absorption rates (OC_{Nitro})** by NSAs also showed some variations in reported literature values, typically within a factor of 2, and largely depending on the specific compound considered (Plumlee & Reinhard, 2007; Shim et al., 2016). Minimum and maximum values of 1.2 and 5 were set for the "worst" and "best" case scenarios, respectively, and the value of 2.5 for the "likely" case. This would correspond to half-lives of 42.8, 85.6 and 178.4 minutes for the "best", "likely", and "worst" case scenarios, respectively. This was in accordance with literature values reported to range from 10-35 min during summer and between 60-220 min during winter (Afzal et al., 2016; Plumlee & Reinhard, 2007; Sørensen et al., 2015). Wind speed can impact the diffusion of compounds between the lake surface and the atmosphere. The wind speed at Lake Elvåga may be significantly lower than the one measured at the meteorological station at Blindern, that was used in the model, due to the lake being situated in a valley with sharp walls of 60-80m. Moreover, the mass transfer velocity, has not been measured for these compounds and is only based on an estimate formula. Due to the impact of NA evasion from the lake water to the air, the diffusive exchange was lowered by a factor of two using a **diffusive exchange multiplier** in the "worst" case scenario compared to the "best" and "likely". The magnitude of the **enthalpy of phase transfer** was completely unknown, and so the same value set for the "worst" case was also applied to the "likely" case scenario. This was done based on the precautionary principle.

Even though the degree of uncertainty associated with some of the scenario parameters was relatively high, the corresponding sensitivity was low for most parameters (≤ 20%). One exception was the biodegradation rate for which the sensitivity was very high (1068%), indicating that the average lake concentration would increase by more than 1000% if this parameter value was changed from its "best" to "worse" case scenario value. This means that the biodegradation rate is the largest cause of uncertainty in the lake NSA and NA concentrations.

3 Results and Discussions

The modelled results are presented by time series (2015 – 2017) of selected key variables of catchment hydrology, lake physics and fluxes of NSA and NA input and degradation for the "likely" case scenario (Figure 2 C-F). Resulting lake water concentrations of NSAs and NAs are presented both separately by time series (Figure 2 A-B) and by the average annual and daily maximum sums for the three scenarios considered (Table 7). See Appendix for time series from the two other scenarios.

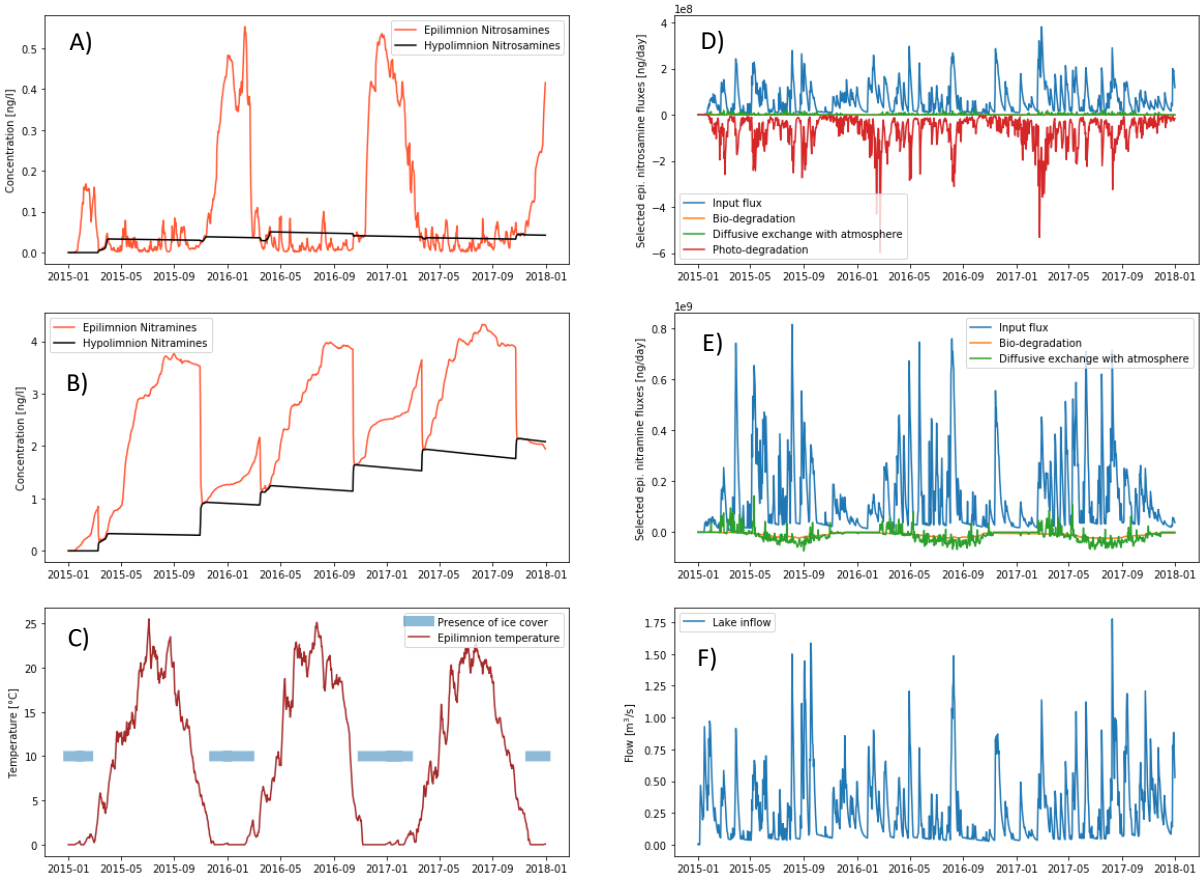

Figure 2: Modelled time series (2015 – 2017) for the "likely" scenario of: A) and B) nitrosamine and nitramine concentrations, respectively, in the top layer of the lake termed epilimnion (orange) and in the bottom layer of the lake termed hypolimnion (black), C) epilimnion lake water temperature (brown) and the presence of ice cover (blue), D) and E) nitrosamine and nitramine fluxes, respectively, of input (blue), biodegradation (orange), diffusive exchange with atmosphere (green), and photodegradation (red), and F) lake water inflow (blue). Rates are in the in order of 0.1 to 1 g day^{-1}.

3.1 Catchment Hydrology and Lake Physics

Overall, the modeling of catchment hydrology and lake physics showed great compliance with data from similar catchments. The catchment model predicted realistic water inflow to the lake (Figure 2 F) with an average flow of 0.28 m^3/s over the period from 2015 to 2017. This was consistent with previously estimated runoff for the same catchment, conducted by NVE using runoff maps (0.35m^3/s: NVE (2006). Particularly when considering the relatively low precipitation in 2016. Moreover, seasonality was evident in the modelled inflow, with the highest flows mainly occurring during precipitation events in autumn. This was in accordance with the seasonality frequently seen for this type of catchment of this region (Vormoor et al., 2015). The lake module simulated realistic lake water temperatures ranging between 0 and 25°C (Figure 2 C). Ice cover was predicted from late November/early December to late February/early March steadily over the three-year period, and with the duration averaging at 98 days. This was consistent with the average ice-cover duration typically reported for small lakes (119 days, lakes < 10 km^2) in the region (Solvang, 2013).

3.2 Rates of NSA and NA Input and Decay, and Resulting Lake Water Concentrations

From the arrangement of the panes in Figure 2 it is clear to see how the seasonal events influences the NSA and NA input and decay rates, and subsequently the lake water concentrations. While the magnitude of the NSAs input fluxes (Figure 2 D) were higher than for the NAs (Figure 2 E), the resulting lake water concentration showed the opposite pattern (Figure 2 A and B). The most important depletion pathway for the NSAs was photodegradation (Figure 2 D), almost balancing out the input flux. The highest NSA lake epilimnion water concentrations were consistently found during winter (Figure 2 A), which coincided with the timing of an ice cover (Figure 2 C), efficiently shielding the NSAs from the moreover weaker sunlight radiation occurring during wintertime. From late spring to early autumn, efficient photodegradation reduced NSA levels to a minimum preventing migration and potential accumulation to the lower lying hypolimnion. The main process influencing the NSA hypolimnion levels was the low biodegradation rate present. Interestingly, the NAs display a different seasonal pattern with a systematic concentration build-up during stratification periods in summer and winter and a strong decrease during lake mixing events (Figure 2 B). The absence of an efficient depletion pathway for the NAs (Figure 2 E) will lead to long-term increases in the hypolimnion and in the lake in general. The peaks in concentration were associated with increased input to the lake during spring snow melt and to a lesser extent during autumn intense precipitation. (For the NSAs, the corresponding higher spring input was mitigated by high rates of photodegradation).

Over the three modelled years (2015-2017), the hypolimnion NA concentration increased from zero to 2 ng L^{-1}. The hypolimnion concentration serves as the baseline for the epilimnion. If the same emission rates continue to persist after the three years, it is very likely that the lake water NA level will continue to increase with time. By running the “likely” scenario for 12 years with the same repeated input data, the epilimnion concentrations stabilize around a yearly mean NA concentration of about 3.7 ng L^{-1}. Note that the background level of NSAs and NAs was set to zero due to the lack of measurements conducted in natural waters.

3.3 Impact of "Best", "Likely", and "Worst" Case Scenarios on Lake Water Concentrations

Daily maximum and annual average epilimnion lake water concentrations for the sum of NSAs and NAs are presented in Table 7 for the three scenarios "best", "likely", and "worst" described in section 2.4. The different scenarios were created to capture the uncertainty of the uncertain key parameters.

Table 7: Annual average (2017) lake water epilimnion concentrations (ng L^{-1}) for the sum of NSAs and NAs modelled for a "best", "likely", and "worst" case scenarios.

	Best	Likely	Worst
Annual average	0.2	3.3	4.4

For the "best", "likely", and "worst" case scenarios, the annual averages (for the year 2017) were 0.24, 3.3, and 4.4 ng L^{-1}, respectively. Thus, the "worst" case scenario was the only to produce concentrations that exceeded the NIPH drinking water limit (4 ng L^{-1}). It should be noted that the best- and worst-case scenarios are considered to be unlikely and only to represent absolute boundary situations in which all the parameter values are simultaneously at extremities of their ranges.

The most important factor for the lower concentrations in the "best" case scenario was a higher biodegradation rate (Table 6). While the photodegradation of NSAs have been quite frequently studied, very few studies have been conducted on the biodegradability of the NAs. Considering the high impact that the biodegradation rate was found to have on lake concentrations, the lack of sufficient studies represents a large uncertainty in the estimated results. Plant uptake should also be studied since it represents another potential depletion pathway.

The result from the "likely" case scenario was compared to the estimate previously made in association with the atmospheric modelling conducted by Norsk Energi (2019). The current average annual "likely" case of NSA and NA lake concentration (3.3 ng L^{-1}) was very close to, but slightly lower than their 3.4 ng L^{-1}. One major difference between the two studies was that our study covered accumulation with time in the stratified lake, which had a large impact on lake NA concentrations.

4 Conclusion

Advanced catchment and lake water modelling simulated lake water concentrations of NSAs and NAs in Lake Elvåga following potential amine-based CC at the nearby waste incineration plant at Klemetsrud. The combined catchment and lake model produced a good fit for the site-specific hydrological and climatic variables which indicates a high degree of site specificity of the simulations.

Catchment and lake processes were found to have a large impact on NSA and NA lake water concentrations and to be highly impacted by seasonal climatic events. While the deposition rates of NSAs were higher than for the NAs, final lake water concentrations after the three-year modelled period showed the opposite picture. Efficient photodegradation of the NSAs was the explanation, almost balancing out the input rates. With the absence of an efficient depletion pathway for the NAs, levels were found to accumulate over time, both in the hypolimnion and epilimnion of the lake.

Seasonal variation had a strong impact on the photodegradation rates of NSAs and on the input rates of both NSAs and NAs. During winter the effect of photodegradation was reduced to a minimum, resulting from the combined effect of weaker sunlight radiation and the presence of an ice cover on the lake. Accordingly, winter-concentrations of NSA were more than five times higher than during the rest of the year. Input rates of both NSAs and NAs were highest during spring, resulting from increased transport to the lake with melting snow. For the NSAs, however, this effect was completely cancelled out by high photodegradation rates during spring.

For the three-year period modelled, maximum annual average concentrations of NSAs and NAs amounted to 0.24, 3.3, and 4.4 ng L^{-1} for the “best”, “likely”, and “worst case scenarios, respectively. These values represent the range of uncertainty in the model result, with values toward the two edges of the interval being less likely than value in the middle. The uncertainty associated with the biodegradation rate was found to be the most important cause of uncertainty in the lake concentrations. The biodegradation rate used in the likely scenario was the one most commonly reported in the literature, while the range used for the other scenarios represent other reported values.

Continued deposition over 10 years seemed to stabilise the annual average value to around 3.5 ng L^{-1}, in the likely case. The high impact from seasonal variation should be taken into consideration for the planning of potential future monitoring programmes. Moreover, a problem related to future monitoring is the lack of sufficiently sensitive methods for the determination of NSAs and NAs in natural waters. In particular, for the NAs, there is no method available to measure concentrations around 4 ng L^{-1} or below. To further reduce the uncertainty of the modelled estimates, future work should focus on assessing the biodegradability of the NAs since this was the parameter found to have the largest impact on the lake NSA and NA concentrations.

5 References

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Appendix

A1 Time Series of the Results from the "best" and "worst" case scenarios.

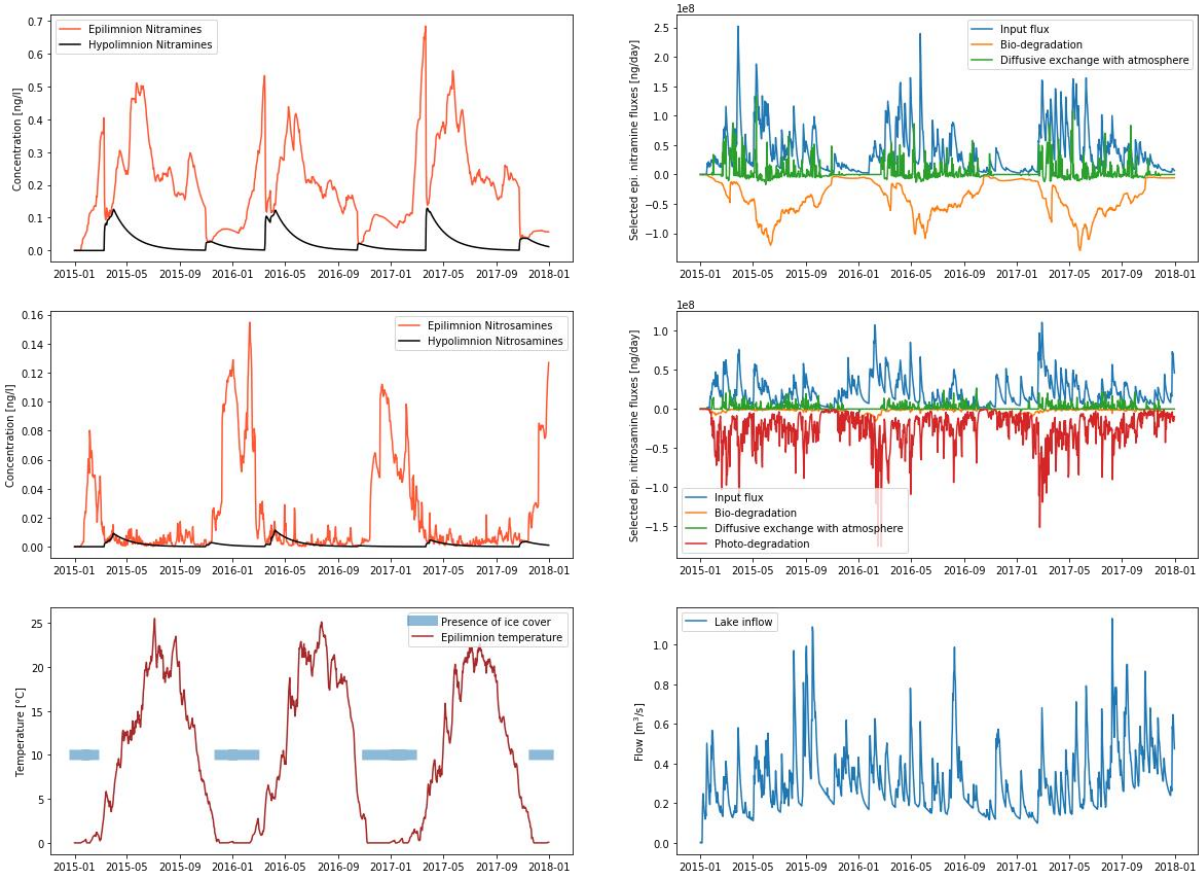

Figure A1: Modelled time series (2015 – 2017) for the "best" scenario of: A) and B) nitrosamine and nitramine concentrations, respectively, in the top layer of the lake termed epilimnion (orange) and in the bottom layer of the lake termed hypolimnion (black), C) epilimnion lake water temperature (brown) and the presence of ice cover (blue), D) and E) nitrosamine and nitramine fluxes, respectively, of input (blue), biodegradation (orange), diffusive exchange with atmosphere (green), and photodegradation (red), and F) lake water inflow (blue). Rates are in the in order of 0.1 to 1 g day^{-1}.

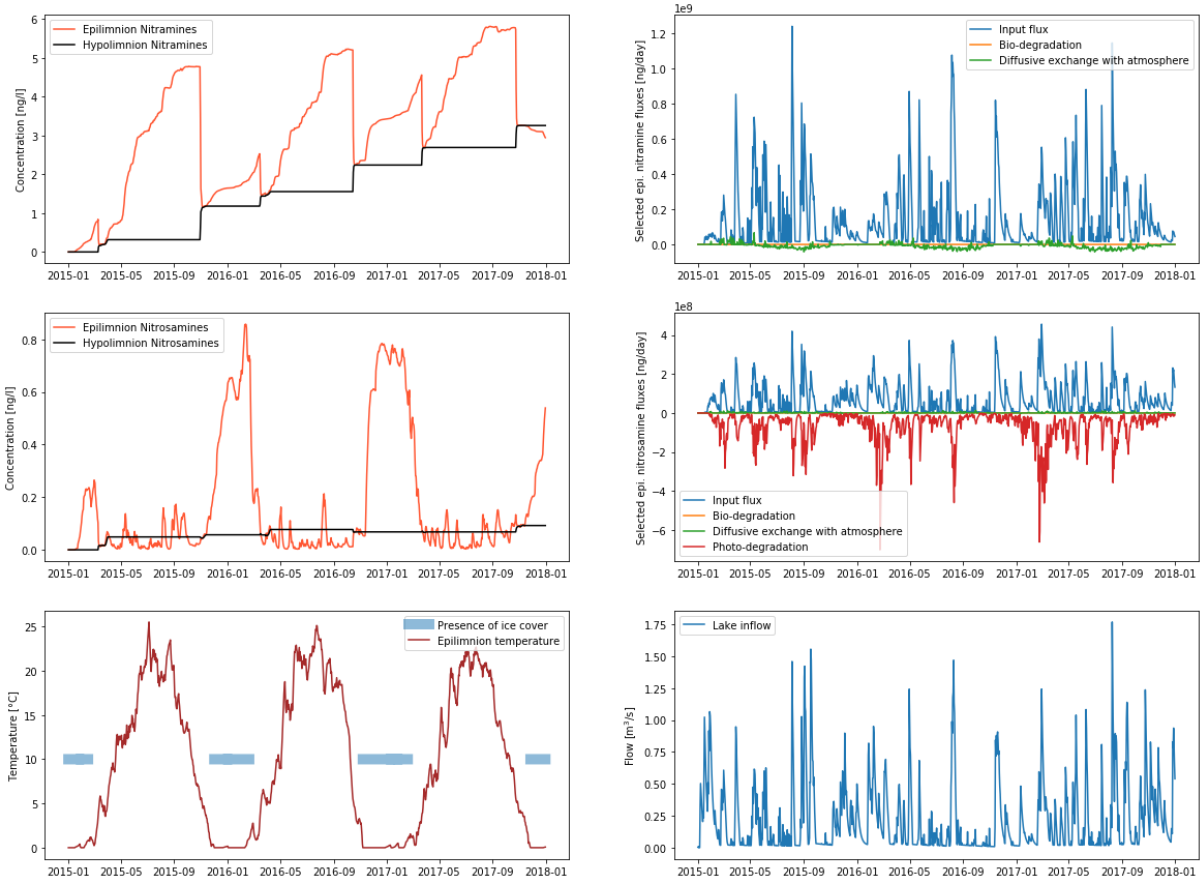

Figure A2: Modelled time series (2015 – 2017) for the "worst" scenario of: A) and B) nitrosamine and nitramine concentrations, respectively, in the top layer of the lake termed epilimnion (orange) and in the bottom layer of the lake termed hypolimnion (black), C) epilimnion lake water temperature (brown) and the presence of ice cover (blue), D) and E) nitrosamine and nitramine fluxes, respectively, of input (blue), biodegradation (orange), diffusive exchange with atmosphere (green), and photodegradation (red), and F) lake water inflow (blue). Rates are in the in order of 0.1 to 1 g day^{-1}.

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