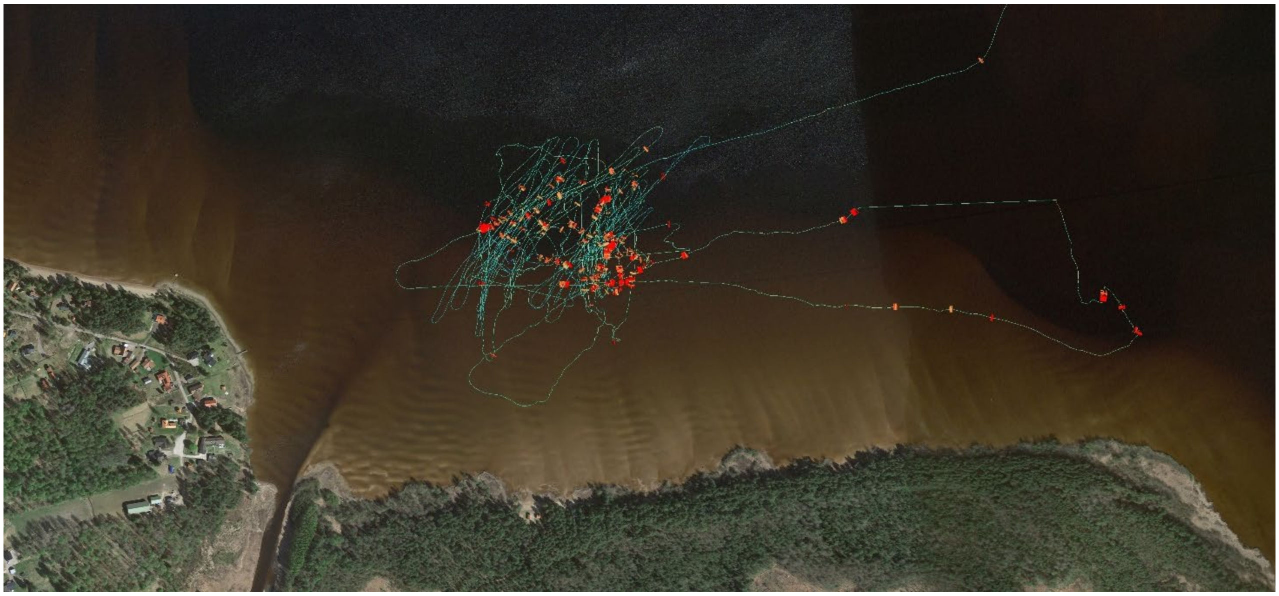
CHALMERS

Measurement of methane leakages from lakes in the Siljan Ring



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Summary

A study was conducted to measure natural methane (CH_4) leaks from lakes in the Siljan Ring—an ancient meteorite crater system where natural gas is present several hundred meters below the surface. The objective was to identify CH_4 emission sources, particularly in areas where local inhabitants have reported recurring winter ice holes. Measurements were carried out during two three-day campaigns, spaced seven months apart, in 2023 and 2024.

This study, for the first time, revealed strong, localized and persistent lake emissions of CH_4 around the Siljan Ring, forming detectable gas plumes in the air (ebullition). Active and highly localized ebullition sources were identified in Lake Orsa (4 locations), Fudalsviken (1 location), and Vikarbyn (2 locations). Elevated concentrations were also observed at Ickviken and around Stumsnäs, both in air and water, although these appear to represent more classical ebullition from lakebed sediment. A few additional potential sources were noted but not followed up. The lakebeds in the region consist mainly of sand, and the leaks were typically found in areas with circulating water and relatively shallow depths (2–5 meters).

Floating chamber measurements indicated CH_4 emissions ranging from 0.05 to 0.2 $\mu g \cdot m^{-2} \cdot s^{-1}$, while carbon dioxide ($CO_2$) emissions were approximately 100 times higher. These diffuse surface CH_4 emissions are up to four times higher than typical ebullition rates observed in lakes. Furthermore, measurements using a novel trace gas method revealed ebullition rates ranging from 20 to 300 g/h—significantly exceeding the diffuse emissions measured by floating chambers and surpassing typical lake ebullition rates by two to three orders of magnitude. This indicates that the studied sources are not only dominated by ebullition but are also exceptionally strong and likely unique. Although the emissions at the hotspot locations are exceptionally strong, their total contribution from a national perspective is negligible. Total CH_4 emissions from the identified hotspots were estimated at approximately 3.5 tons per year (equivalent to 85 tons of CO_2), with additional diffuse emissions contributing around 50 kg per year (1 ton CO_2 equivalent). These emissions represent a very small fraction—about 20 parts per million—of Sweden's total CH_4 emissions, according to the Swedish National Greenhouse Gas Inventory. However, this study only targeted hotspots along the eastern shores of Lake Siljan and the southern part of Lake Orsa, and the findings may represent just the tip of the iceberg. Many more sources may exist along other parts of the lakes, in nearby water bodies, at greater depths, or with lower but still significant emission strengths.

Due to the limited number of measurements and potential sampling artifacts, it remains unclear whether the gas is of thermogenic or biogenic origin. Further investigation, including detailed analysis of VOC content and $\delta^{13}C$ isotopic composition, is needed.

The emission sources in the Siljan Ring are unique in being localized, persistent and in source strength —an uncommon characteristic for lake ebullition, which typically occurs sporadically over broad areas with emission several order of magnitude lower. These types of emissions are difficult to detect and quantify using traditional methods, such as floating chambers or eddy covariance systems.

Further research is needed to assess temporal variability, identify additional sources in the Siljan Ring (which may be numerous), and evaluate their impact on Sweden's natural climate budget. Additionally, similar lake systems elsewhere should be investigated, and mitigation strategies may need to be developed.

Sammanfattning

En studie har utförts för att mäta naturliga metanläckage (CH$_4$) från sjöar i Siljansringen – ett gammalt meteoritkratersystem där naturgas finns på flera hundra meters djup. Målsättningen var att identifiera förekomsten av CH$_4$-utsläppskällor, särskilt vid platser där lokala invånare rapporterat återkommande isvakar under vintern. Mätningarna genomfördes under två tredagarskampanjer, med sju månaders mellanrum, under 2023 och 2024.

Denna studie visade, för första gången, starka, lokalt avgränsade och ihållande metanutsläpp från sjöar runt Siljansringen, som bildade detekterbara gasplymer i luften genom så kallat bubbelutsläpp (ebullition). Aktiva och mycket lokala bubbelkällor identifierades i Orsasjön (4 platser), Fudalsviken (1 plats) och Vikarbyn (2 platser). Förhöjda koncentrationer observerades även vid Ickviken och kring Stumsnäs, både i luft och vatten, men dessa förefaller representera mer klassiska bubbelutsläpp från bottensediment. Några ytterligare potentiella källor noterades men följdes inte upp. Sjöbotten i området består huvudsakligen av sand, och de observerade utsläppen återfanns på platser med cirkulerande vatten och vid relativt grunt djup (2–5 meter). Mätningar med flytande kammare visade CH$_4$-utsläpp i storleksordningen 0,05–0,2 $\mu g \cdot m^{-2} \cdot s^{-1}$, medan koldioxid (CO$_2$) släpptes ut i ungefär 100 gånger större mängder. Dessa diffusa metanutsläpp från vattenytan är upp till fyra gånger högre än typiska bubbelutsläpp från svenska sjöar. Därtill visade mätningar med en ny typ av spårgasteknik att bubbelutsläppen kunde uppgå till 20–300 g/h – avsevärt högre än de diffusa utsläpp som mättes med flytkammare, och två till tre storleksordningar högre än vad som är normalt för sjöar. Detta visar att de studerade källorna inte bara domineras av bubbelutsläpp, utan också är exceptionellt starka och sannolikt unika.

Även om utsläppen från dessa s.k. ”hotspots” är mycket kraftiga lokalt, är deras totala bidrag ur ett nationellt perspektiv försumbart. De totala CH$_4$-utsläppen från de identifierade hotspotsen uppskattades till cirka 3,5 ton per år (motsvarande 85 ton CO$_2$-ekvivalenter), med ytterligare diffusa utsläpp motsvarande cirka 50 kg per år (1 ton CO$_2$-ekvivalenter). Tillsammans utgör dessa utsläpp endast omkring 20 miljondelar av Sveriges totala CH$_4$-utsläpp, enligt den svenska växthusgasinventeringen. Det är dock viktigt att notera att studien enbart fokuserade på hotspots längs de östra stränderna av Siljan och den södra delen av Orsasjön. Våra fynd kan alltså bara utgöra toppen av ett isberg. Det är troligt att fler utsläppskällor finns runt andra delar av sjöarna, i närliggande vattendrag, på större djup eller med lägre men ändå betydande utsläppsnivåer. På grund av det begränsade antalet mätningar och möjliga provtagningsfel är det fortfarande oklart om metanet har ett fossilt (termogent) eller biologiskt (biogent) ursprung. Vidare undersökningar krävs, inklusive detaljerad analys av VOC-innehåll och isotopsammansättning ($\delta^{13}C$).

Utsläppskällorna i Siljansringen är unika genom att de är både lokalt avgränsade och långvariga och lokalt väldigt kraftiga – något som är ovanligt för bubbelutsläpp i sjöar, vilka normalt förekommer mer slumpmässigt över större områden. Dessa typer av utsläpp är svåra att upptäcka och kvantifiera med traditionella metoder, som flytkammare och eddy-kovarianssystem. Vidare forskning behövs för att undersöka variationen i utsläppen över tid, identifiera fler utsläppskällor i Siljansringen (vilka kan vara många) samt utvärdera deras påverkan på Sveriges naturliga klimatbudget. Även andra sjösystem bör studeras, och det kan bli aktuellt att utveckla strategier för att hantera eller mildra dessa utsläpp.

Abbreviations

CH_4 – methane

CO_2 – carbon dioxide

C_2H_6- ethane

TDM – Tracer Dispersion Method

FTIR – Fourier Transform Infrared (Spectroscopy)

NDACC – Network for the Detection of Atmospheric Composition Change

ICOS – Integrated Carbon Observation System

VOC – Volatile Organic Compounds

$\delta^{13}C$ – Carbon Isotope Ratio (Delta-13 Carbon)

CEN – European Committee for Standardization

ppb – Parts Per Billion

$\mu g \cdot m^{-2} \cdot s^{-1}$ – Micrograms Per Square Meter Per Second

g/h – Grams Per Hour

ppm – Parts Per Million

GWP – Global Warming Potential

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1. Introduction

Open water lake emissions have been identified as the second largest global natural source of methane (CH_4) (1), but flux estimates vary by a factor of 5 (2,3,4). CH_4 can be emitted from lakes to the atmosphere via several pathways, including diffusion, ebullition (bubble emission), release from storage, and flux mediated by emergent plants (4). Diffusive flux depends partly on the CH_4 concentration gradient between the water and the air, and on the turbulence in the surface water, which influences gas transfer rates. Ebullition is highly dependent on CH_4 production in the sediments and on physical factors triggering bubble release and exhibits high spatiotemporal variability. Diffusive fluxes can be calculated from concentration measurements of degassed water samples using headspace analysis with instruments such as gas chromatographs or laser-based spectrometers (5) or measured directly via eddy covariance (6) or floating chambers (7). In a recent study, CH_4 emissions were measured from four Swedish lakes with different physicochemical characteristics using floating chambers, showing diffuse emissions ranging between 0.02–0.05 $\mu g\ m^{-2}\ s^{-1}$, and a strong pattern over day and night (8). Ebullition is more difficult to assess, due to the large spatiotemporal variability although this process is claimed to dominate emissions into the atmosphere in stratified waters (9, 10, 11). The study area, the Siljan Ring (Figure 1), is a meteorite crater with deep faults extending up to 20 km deep across hundreds of kilometers. Oil was produced in the Rättvik area for industrial use in the 19th century, and seep oil has been observed for a long time (12). The fault system currently hosts several lakes, with Lake Siljan, the largest, located on the SSW edge, and Lake Orsa to the west. Natural gas prospecting has focused on extracting gas from fractured crystalline bedrock beneath 200–600 m thick sedimentary caprock, with boreholes drilled to 400–700 m depths (13). CH_4 accumulations have been detected both in the caprock and the granite fracture system. The west side of the crater shows more gas, while oil is more common on the east side, along with small hydrocarbon levels. CH_4 can form through three mechanisms in the deep subsurface: abiotic, via inorganic reactions, e.g., H_2 and carbon dioxide (CO_2), typically through magmatic processes; thermogenic, from organic matter breakdown at high temperatures; and biogenic, through microbial activity. Thermogenic CH_4 is often associated with heavier alkanes, such as ethane and propane. A recent study (14) claims the existence of microbial methanogenesis and CH_4 oxidation deep within the fracture system, with evidence of fossilized anaerobic fungi together with methanogen bacteria (15). Data from boreholes indicate that CH_4 is largely biogenic but mixed with thermogenic CH_4. Seep oil and bitumen in limestone likely originate from organically rich shale. Recent more advanced analysis of the borehole data (22) indicates that the majority of CH_4 in this system is predominantly generated by microbial activity. This conclusion is derived from the specific carbon isotope signatures of $\delta^{13}C$ in CH_4, which are highly negative (-57 ‰ to -66 ‰), and the slightly negative to positive $\delta^{13}C$ in CO_2 (-14 ‰ to 6 ‰). Additionally, the authors suggest that some CH_4 may have a thermogenic origin, produced deep underground through the heat-driven breakdown of organic matter. This hypothesis is supported by the presence of heavier hydrocarbons, such as ethane and propane, which are typically associated with thermogenic processes.

The aim of this pilot study at the Siljan Ring was initially to investigate whether there is evidence of deep CH_4 leaking through geological fissures. In this study, we tested the hypothesis that persistent winter ice holes, as reported by the local population (pers. comm. Mats Budh), are associated with CH_4 leakages. Measurements were conducted during two 3-day campaigns, separated by six months, in the fall of 2023 and early summer of 2024. In the second campaign a general mapping along the SE shore of Siljan was also carried out.

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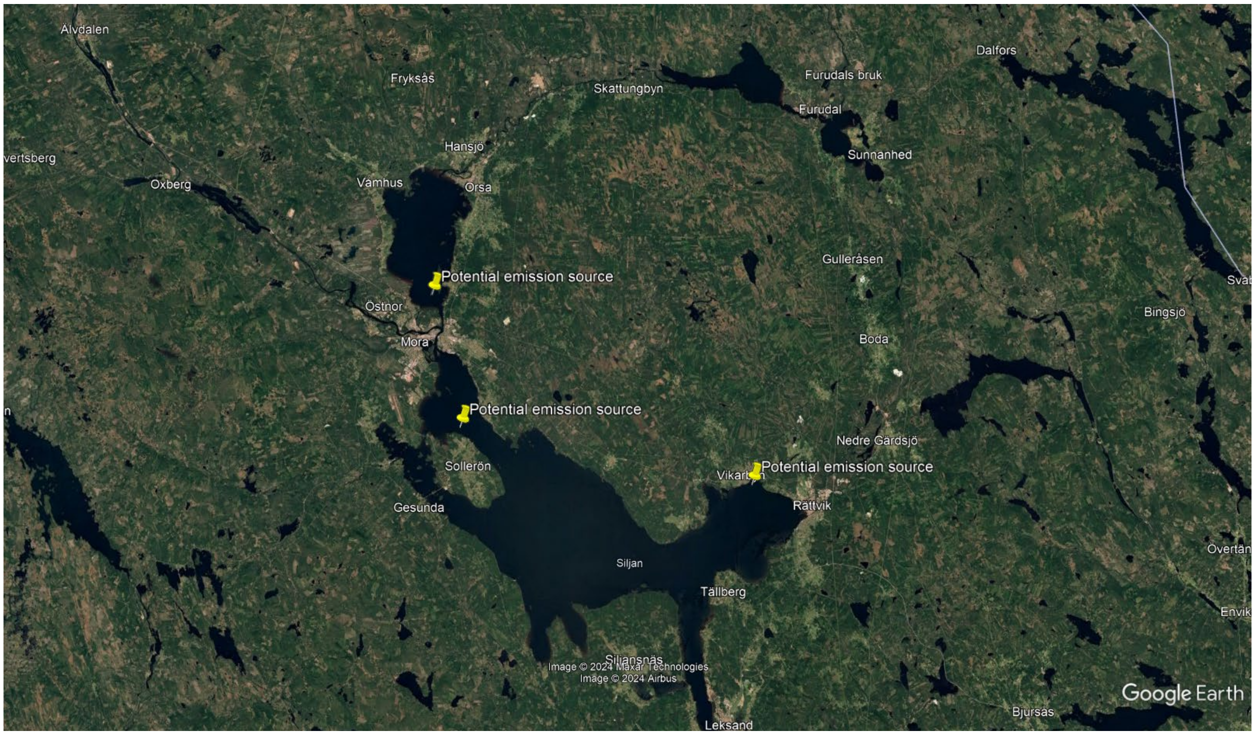

Figure 1. Overview of the Siljanring, a crater system caused by implosion of a meteroid 377 M years ago. Some potential CH_4 emission hotspots are shown.

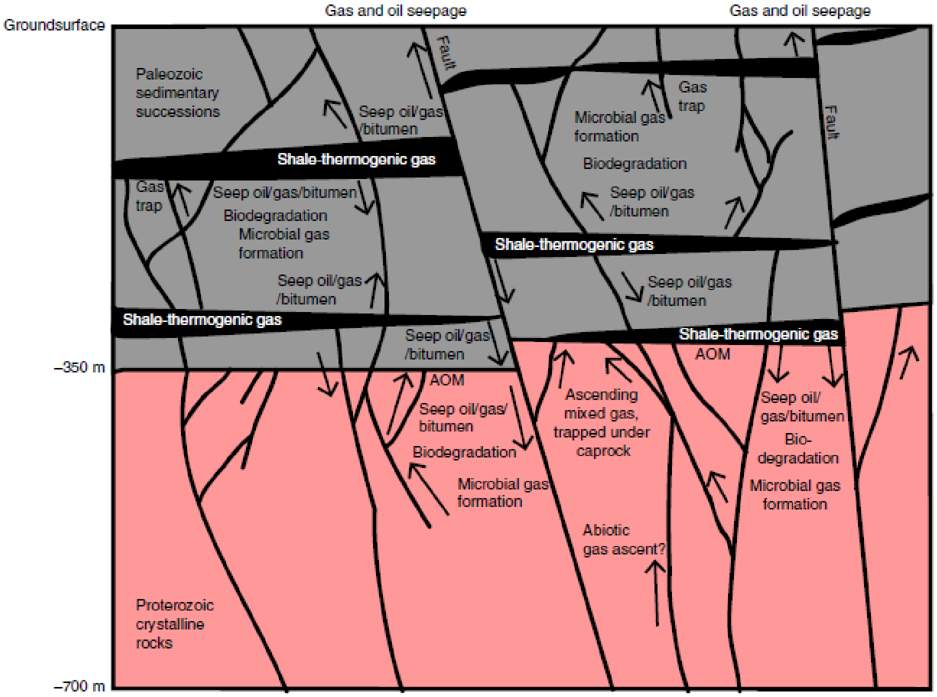

Figure 2. The leakages of CH_4 and heavier VOCs at the Siljan ring may be due to cracks and faults in a caprock layer with shale inclusions, preventing gas diffusion except in these areas. The lake's leakages likely stem from deep faults or weaker caprock. CH_4 forms from both biogenic and thermogenic sources, along with various VOCs. Adopted from (13).

2. Method

2.1 Instrument description

Three types of laser sensors were employed to measure CH_4, ethane (C_2H_6), propane (C_3H_8), and CO_2, offering high sensitivities and a time response of 1 second (Figure 3). CH_4 and CO_2 were measured using a Picarro G2301 cavity ring-down spectrometer with an accuracy of a few hundred ppb for CO_2 and a few ppb for CH_4. Ethane and propane were measured using two different Aeris Strato instruments, with an accuracy of approximately 2 ppb for ethane and 5–10 ppb for propane. These sensors provided real-time, high-resolution measurements, making them suitable for capturing transient plumes and small-scale spatial variations in gas concentrations. It should be noted that propane was only measured in the first campaign.

The Picarro sensor is the backbone of the global CO_2 and CH_4 monitoring networks (e.g., http://www.icos-sweden.se), while the smaller, lighter Aeris sensor is increasingly used for mobile and airborne applications. Although these laser sensors do not require traditional calibration, CH_4- and CO_2-measurements were validated using calibration gases with an accuracy of ±0.5-5%. Ethane and propane measurements were validated against a Fourier-transform infrared (FTIR) spectrometer at the Chalmers laboratory.

An additional laser sensor, the Picarro G2201-i isotope analyzer, was used to measure isotopic ratios ($\delta^{13}C$) in CO_2 and CH_4, enabling source apportionment and isotopic signature analysis. This instrument achieves isotopic precision of ±0.5‰ for CH_4 and ±0.2‰ for CO_2. During the measurement campaign, raw water samples were collected in gas-tight bottles, and the gas in the headspace was subsequently analyzed to complement the airborne and mobile monitoring efforts.

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Figure 3. Two Aeris laser sensors (Aeris Strato) were used to measure methane, ethane, and propane, with a sensitivity of 1 ppb and a time resolution of 1 second. An additional sensor, the Picarro G2301, was used to measure carbon dioxide (CO_2) with a precision of 200 ppb. A third laser sensor, the Picarro G2201-i isotope analyzer, was used in the laboratory. This instrument combines the capabilities of two Picarro $\delta^{13}C$ carbon isotope instruments for CO_2 and CH_4 into a single unit.

2.2 Floating chamber

The floating chamber method measures vertical gas fluxes, such as CH_4 and CO_2, from water and soil surfaces to the atmosphere (Figure 4). A floating chamber placed over the water surface captures gas exchange, with flux determined by changes in gas concentration over time. In this study, a Picarro G2301 gas analyzer was used, with both inlet and outlet connected to the measurement chamber. The analyzer continuously circulated the air through the chamber and measured the buildup of CH_4 and CO_2 concentrations. The flux is calculated using the rate of change in concentration (Eq. 1).

$$dC(t)/dt = E/V - k \cdot C(t) \qquad \text{(Eq. 1)}$$

where E is the emission rate, V the chamber volume, k the leakage constant, and C(t) the concentration at time t. The solution is:

$$C(t) = (E / (k \cdot V)) \cdot (1 - \exp(-k \cdot t)) + C(0) \cdot \exp(-k \cdot t) \qquad \text{(Eq. 2)}$$

At equilibrium ($t \to \infty$), the concentration is:

$$C_{\text{equilibrium}} = E / (k \cdot V) \qquad \text{(Eq. 3)}$$

Thus, the emission rate can be calculated as:

$$E = k \cdot V \cdot C_{\text{equilibrium}} \qquad \text{(Eq. 4)}$$

On June 27, the system functioned without leakage (k=0). On June 25 and 26, however, pump leakage (~0.9 L/min) led to an underestimation of emissions, particularly affecting CO_2 results, which were excluded for these days. CH_4 data seemed satisfactory but was marked as uncertain due to potential underestimation.

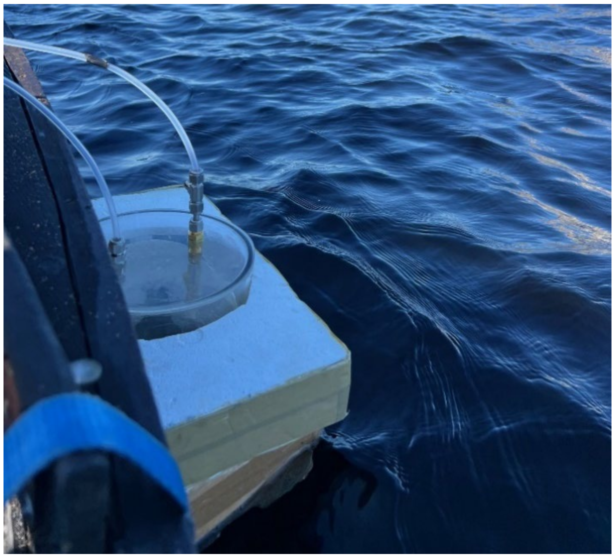

Figure 4. The floating chamber is placed over the water surface, where the gas exchange between the water surface and the atmosphere is captured within the chamber. This allows for determining the gas flux by measuring the change in gas concentration inside the chamber over time. In this study, the chamber was connected to a gas analyzer (Picarro), with the sampling air continuously recirculated through the chamber while measuring the concentration of CH_4 or carbon dioxide in the chamber air. The change in gas concentration over time in the chamber is used to calculate the gas flux.

2.3 Tracer dispersion measurements

The tracer dispersion method (TDM), also known as the tracer correlation method (TC), is used to quantify gas emissions from identifiable point sources. TDM has been standardized by CEN (European Committee for Standardization) since 2021 (16) for quantifying gas emissions from various sources in refineries and chemical plants (17). Based on research by the Chalmers group (18, 19, 20), TDM has been applied in numerous field measurements, e.g. at landfills, industrial facilities, and gas wells.

TDM is based on releasing trace gas (tracer) with a controlled rate near the actual leakage source. It is assumed that both the tracer and the source gas undergo similar dispersion and dilution. When applying TDM, the tracer is released near the source, and the concentration of the trace gas across the plume over a certain path (from x_1 to x_2) is measured.

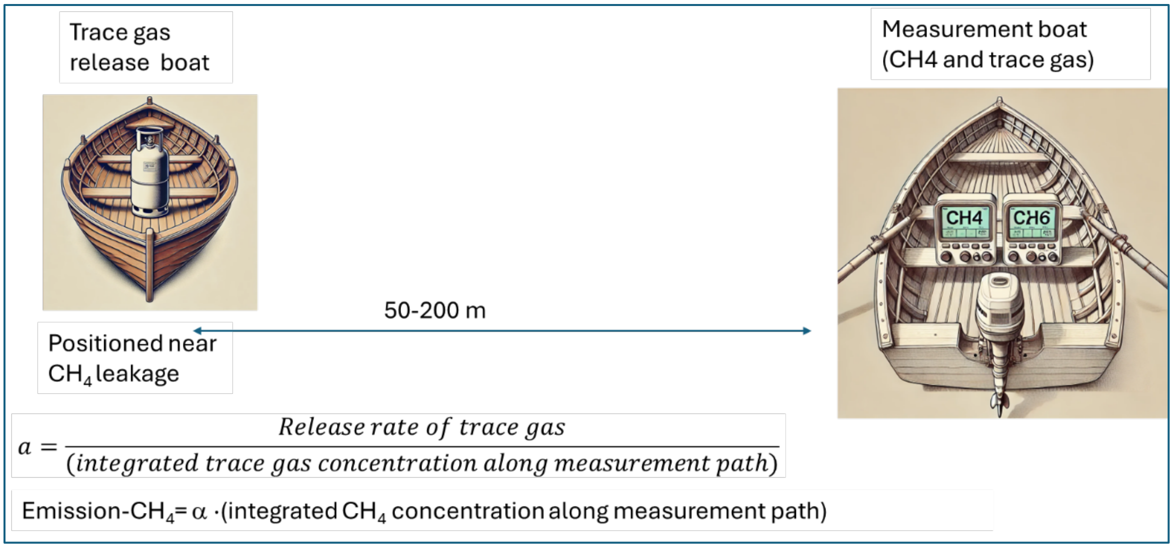

Figure 5. Illustration of a tracer dispersion measurement. The trace gas, ethane, is released in a controlled manner from a measurement boat in the vicinity of an identified CH_4 source. The ratio between CH_4 and ethane is then measured, integrated over the plume, approximately 50–200 meters downwind. The ratio between the trace gas release rate and the integrated amount along the measurement path provides the dilution factor. This can then be used to calculate the CH_4 emissions from the source.

The dilution factor α is calculated as the ratio between the path-integrated trace gas concentration and its release rate, as shown in Eq. 5. Since both source gas and tracer are dispersed similarly, the dilution factor α also applies to the source gas. Emissions are then calculated by integrating the source gas concentration, measured simultaneously with the trace gas, across the plume and multiplying by the dilution factor α, as illustrated in Figure 5 and Eq. 6. In this study, CH_4 was used as the source gas and ethane as the trace gas. The release rate of the trace gas was obtained by weighing the gas cylinders and recording the release time. An example from one of the identified leakage sources at Vikarbyn is shown in Figure 6.

$$a = \frac{Qtracer}{\int_{x1}^{X2} C_{tracer}\, dx} \qquad \text{(Eq. 5)}$$

$$E_{source} = a \cdot \int_{x_1}^{x_2} C_{source}\, dx \qquad \text{(Eq. 6)}$$

Here Q_{tracer} corresponds to release rate of trace gas (kg/h), E_{source} corresponds to emission rate of the source gas and C_{tracer} and C_{source} correspond to the measured concentration of tracer and source gas.

To ensure the trace gas accurately represents the source emissions, it is essential to release the trace gas near the emission source, which was determined by a detailed concentration mapping around the source area. Multiple measurements under varying conditions and distances are conducted to minimize uncertainties and improve accuracy.

Figure 6. Ethane was utilized as a trace gas and released in a controlled manner near the identified CH_4 leakage sources. This figure shows measurements taken at Vikarbyn, southern Siljan, where the concentrations of CH_4 and ethane were measured approximately 50 meters downstream.

2.4 Isotope analysis and ethane-to-CH_4 ratio.

The isotopic composition of carbon in CH_4 varies by origin. Biogenic CH_4 has lower $\delta^{13}C$ values (−60‰ to −50‰) due to microbial processes, while thermogenic CH_4 shows less negative $\delta^{13}C$ values (around −40‰). CO_2 from biogenic CH_4 oxidation has $\delta^{13}C$ values ranging from −30‰ to +10‰, depending on fractionation. Analyzing $\delta^{13}C$ in CH_4 from lakes and boreholes can reveal common origins. Isotope measurements were done with a Picarro G2131-i analyzer.
Additionally, the ethane-to-CH_4 ratio indicates origin: biogenic CH_4 lacks ethane, whereas thermogenic CH_4 contains 1–5% v/v ethane.

A recent study found $\delta^{13}C$ in CH_4 from three test boreholes at the Mora airfield to be −65.8‰, −63.2‰, and −58.8‰ for Boreholes 1, 2, and 4, respectively. CH_4 origins were mixed, with Borehole 1 showing more thermogenic influence and Boreholes 2 and 4 indicating a microbial origin. The δD and $\delta^{13}C$ values in CH_4 supported a primary microbial origin. CO_2 isotopes ($\delta^{13}C$) ranged from −13.8‰ in Borehole 4 to +5.5‰ and +6.4‰ in Boreholes 1 and 2, consistent with microbial processes like carbonate reduction.

3. Experimental

In this pilot study at Lake Siljan and Lake Orsa, the focus was on studying CH_4 emissions in locations around the Siljan Ring where persistent winter ice holes are often reported, according to local knowledge (Figure 7). The hypothesis is that these ice holes could be associated with CH_4 leaking through geological fissures from larger depths.

Measurements were carried out on two 3-day campaigns separated by an interval of six months, in the fall of 2023 and early summer of 2024. Ebullition rates (bubbling emissions) were estimated using a variant of the Chalmers-developed tracer dispersion technique, while diffusive emission rates near hotspots and in background areas were measured with floating chambers.

During the first campaign, two Aeris Strato laser sensors were used to measure CH_4, ethane, and propane. The instrumentation was placed in an aluminum box and carried on several small boats and occasionally hand-carried for performing land-based measurements (Figure 10). The laser sensors could sample ambient air directly or from a floating chamber that was occasionally deployed (Figure 4). This first study covered several areas, both on land and water including a test drilling site at Mora Airfield and a woodchip thermal combustion plant. Measurements were also carried out in southern part of Lake Orsa and northeast shores of Lake Siljan.

During the second campaign in June 2024, the geographical scope was expanded to include mapping of CH_4 hotspots along a considerable part of the eastern side of Siljan in addition to more detailed studies at the southern part of Lake Orsa. The objective was also broadened to include quantification of emissions using trace gas measurements and floating chambers. In addition, several water samples were also taken for isotopic analysis at the University of Gothenburg. The later type of measurements was not planned, and there are potential measurement artifacts due to sample handling.

During the second campaign a slightly larger measurement boat (Figure 8) was used which was equipped with a Picarro instrument measuring CO_2 and CH_4 and an Aeris Strato sensor measuring CH_4 and ethane. Ambient air was drawn to the instruments through Teflon tubing connected to a 1.5 m mast on the boat. The inlet and outlet of the Picarro were also connected to a floating chamber on occasions. A pump leakage occurred during some of the experiments making some of the measurements uncertain as discussed below. An additional dinghy was used to carry the tracer gas setup, as shown in Figure 6.

A 20-liter gas cylinder containing pure ethane was used as a trace gas. A gas regulator, connected to a flow meter on the cylinder, controlled the release. The release timing was recorded, and the cylinder's weight was measured before and after releases. The flowmeter was manually checked periodically to ensure a steady flow.

Figure 7. An ice wake is shown on Lake Siljan, NE of Sollerön, which often occurs at the same location (Photo by Mats Budh). According to the local population, numerous such wakes form on the ice of lakes within the Siljan Ring during winter, including Siljan, Lake Orsa, Skattungsjön, and Oresjön, with variations in size. This study assumed that these wakes may be caused by CH_4 ebullition. Three known wakes are marked as blue points in Figure 1, and at two of these locations, emissions of CH_4 were detected during the prestudy.

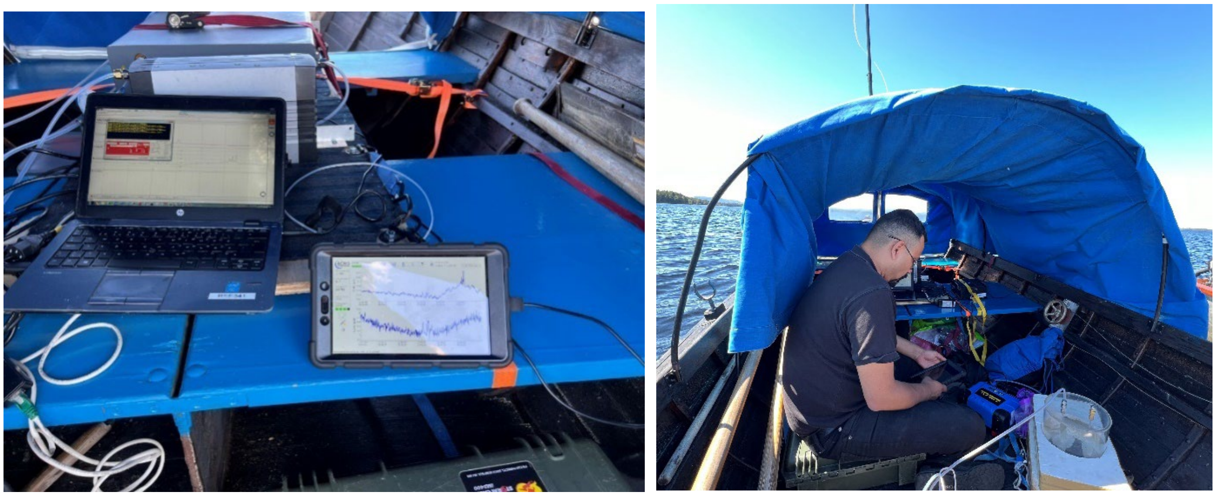

Figure 8. The experimental setup used during the campaign. On the left, laser sensors and real-time displays are shown in the foreground. On the right, the measurement boat is shown—a seven-meter-long wooden vessel with a gas intake mounted on a 1.5-meter mast at the front, connected to the analyzers in the left image.

4. Results

4.1 Mapping 2023

In October 2023, a mapping survey in the Siljan Ring area detected elevated CH_4 and ethane near Mora Airfield due to a borehole leak (Figure 9). The observed ethane-to-methane ratio was 1 % which is consistent with van Dam (22) and which indicates thermogenic fossil fuel origin. Additional small amounts of these gases were found near other boreholes and a thermal wood combustion plant. During the first campaign, mobile measurements at Lake Siljan (Figure 10), north of Sollerön, revealed higher CH_4 concentrations in water south of a small island, with minimal ebullition observed except for one high concentration instance. At Lake Orsa, persistent elevated CH_4 levels (+100 ppb) were recorded over several hours, suggesting an ebullition hotspot. Floating chamber measurements confirmed higher CH_4 in water compared to air. No ethane was detected; however, even assuming a 1% ethane-to-methane ratio, the expected ethane levels would be below the detection limit given the generally measured CH_4 concentrations. This pre-study highlighted the presence of persistent ebullition in Lake Orsa, necessitating further measurement and quantification to understand this phenomenon better.

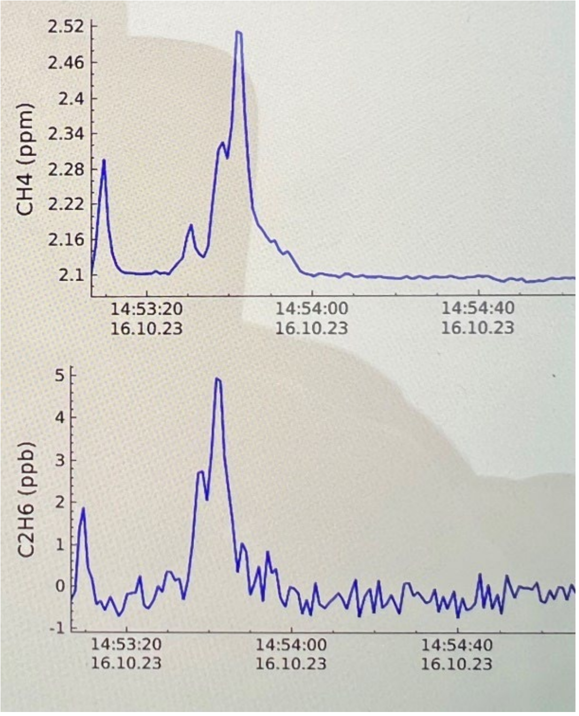

Figure 9. Measurement of CH_4 and ethane leakage from a 450 m deep test borehole showed ethane at 1% relative to CH_4, indicating thermogenic origin.

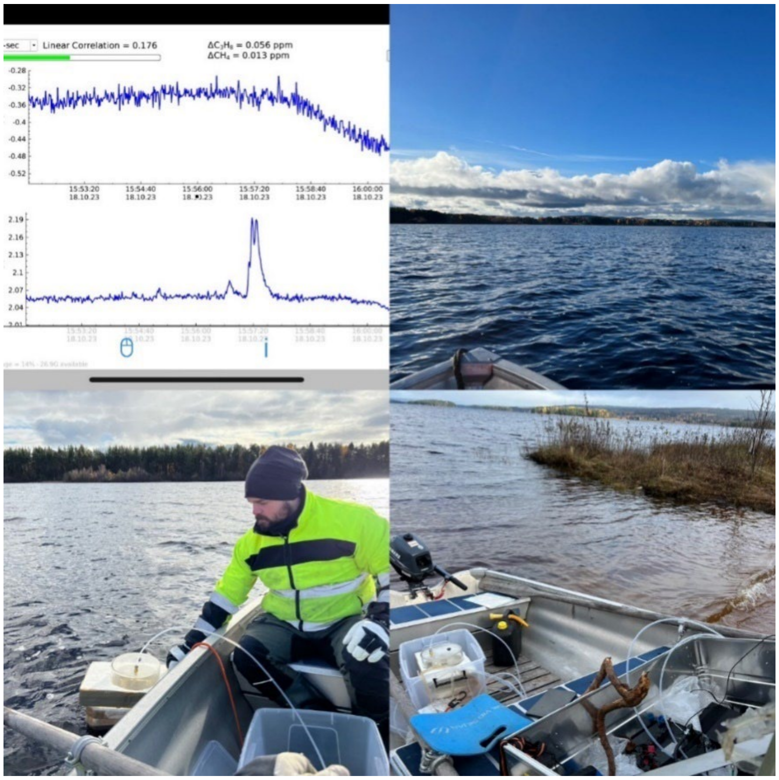

Figure 10. Measurement at Lake Orsa on October 17, 2023. Distinct CH_4 plumes exceeding 100 ppb were observed in the air over a 100 x 100 m area in the southern part of Lake Orsa. In the upper left corner examples of CH_4 (lower graph) and ethane (upper graph) concentrations measured in the air are shown.

4.2 Mapping 2024

During a few days in June 2024, a second campaign was conducted to build upon the findings from the 2023 pre-study and expand the scope to quantify emissions. As a first step, air concentration mapping was carried out near the eastern shores of Siljan Lake and the southern bay of Lake Orsa. The measurements were conducted using the setup shown in Figure 8, focusing on investigating whether CH_4 emissions were associated with areas of reported persistent ice wakes. The results are displayed in several figures, with red color indicating highly elevated levels of CH_4, known as CH_4 hotspots.

Figure 11 shows CH_4 concentrations near the northeastern shores of Lake Siljan on June 25. A distinct source (Figure 12) was observed both in the morning and afternoon in Fudalsviken, near the harbor inlet, at depths of 4 meters or more. This CH_4 source was located just south of two artificial islands acting as piers, with emissions appearing to originate from a depth of at least 5 meters. Most measurements were taken approximately 50-100 meters downwind, to the north of the piers. The location of this source was located to about 30 m and there were elevated levels in the water (see sections below). Elevated CH_4 levels were also observed north of Utanmyra on Sollerön, but these concentrations were intermittent and were therefore not further investigated during this campaign.

Figure 11. CH_4 concentration mapping in the air near the northeastern shores of Lake Siljan on June 25, 2024. A distinct CH_4 source was observed at Fudalsviken, near the harbor inlet, originating from a depth of 5 meters or more. Elevated concentrations were also detected north of Utanmyra on Sollerön.

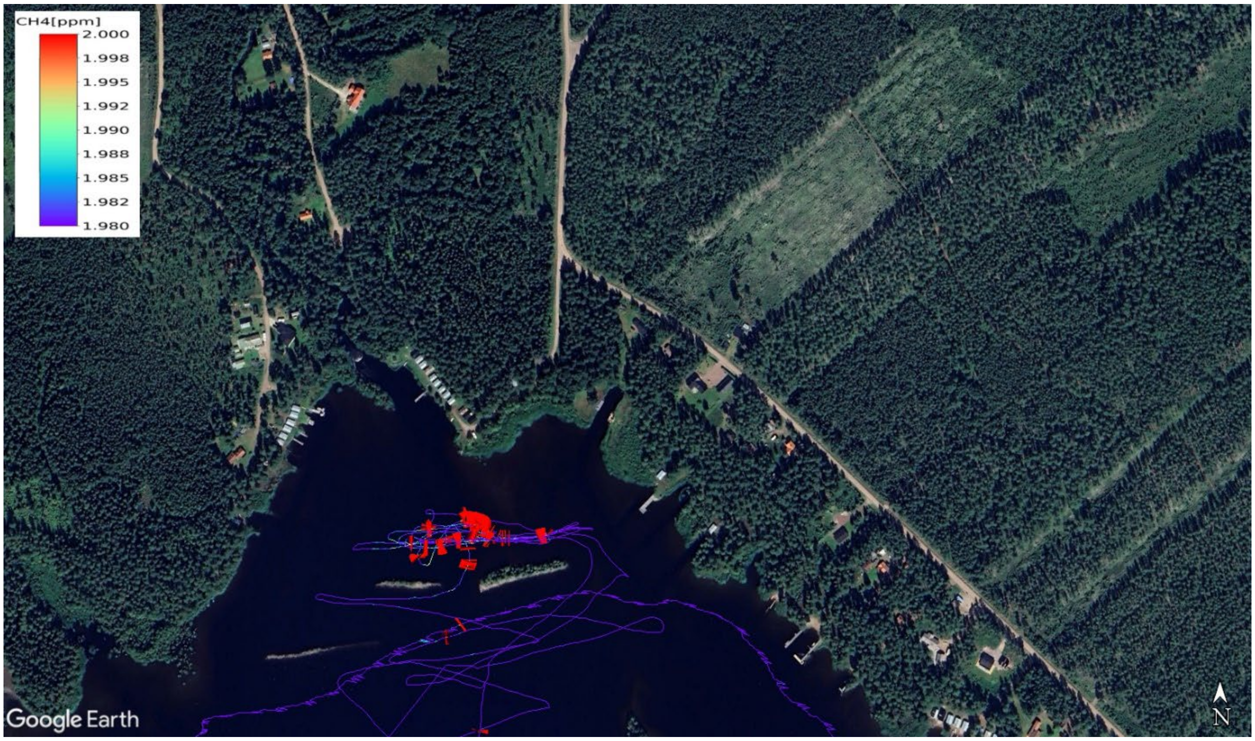

Figure 12. CH_4 concentration mapping in Fudalsviken on June 25, 2024. A distinct CH_4 source was observed near the harbor inlet, just south of two artificial islands acting as piers, with emissions originating from a depth of 5 meters or more. Most measurements were conducted approximately 50 meters downwind, north of the piers.

Figure 13 shows atmospheric CH_4 concentration near southeastern Siljan shores on June 26. Multiple hotspots were observed, with some investigated in detail and others not due to time constraints. The far-right source (Figure 14) is located in Åkernäsviken at Vikarbyn at shallow depth (1 m) and was localized for several minutes, suggesting it may not be typical ebullition from sediments. The second source from the right, is on top of the old foundation for an old steamboat (Figure 14 and Figure 15), and it was studied for an hour and quantified using TDM. It appeared highly localized and persistent, localized within an area of 10 by 10 m, indicating emissions from greater depths. Bottom material in these areas was primarily sand. Disturbing the sediment near the hotspot with a rowing oar increased CH_4 levels, suggesting the sand was saturated with CH_4. No other ebullition was observed along the shores of Vikarbyn. Since the lakebed appeared similar throughout the area, this supports the hypothesis that the emission originates from a deeper source rather than from shallow sediment.

An additional CH_4 hotspot—the third from the right in Figure 13—was found in Ickviken. Two more CH_4 hotspots were observed southwest of Stumsnäs. These latter sources were located in shallow waters with more visible sediment and vegetation (including reed beds) and likely represent typical ebullition form degradation of sediments in the lakebed, as their spatial localization varied significantly.

Figure 13. CH_4 concentration mapping near the southeastern shores of Lake Siljan on June 26, 2024. Higher concentrations (shown in red) were identified at two point sources in Vikarbyn—Åkernäsviken and an old steamboat pier—and more classical ebullition sources from shallow sediments around Ickviken (the large bay in the center) and at two locations near Stumsnäs.

Figure 14. CH_4 concentration mapping at two sources in Vikarbyn: old steamboat pier (left) and Åkersnäsviken (right).

Figure 15. Hotspot area r in Vikarbyn, showing localized CH_4 emissions observed at shallow depth. This source was persistent over several hours and located atop rocks forming the foundation of a former steamboat pier.

Figure 16 shows atmospheric CH_4 concentration mapping over Lake Orsa on June 27, 2024. Two clearly identifiable sources, each approximately 10 × 10 meters in size, were located at depths of 2–3 meters and exhibited persistent ebullition. The bottom material in these areas consisted primarily of sand. Disturbing the sediment near the hotspots with a rowing oar caused significant CH_4 spikes, indicating that the sand was saturated with methane.

Additionally, at least two smaller sources were present in the same area. All these sources were also observed during the first campaign in October 2023. In this region, we also measured diffuse CH_4 emissions from the water surface and collected water samples for isotopic analysis (see next sections). The persistence and strong localization of these emissions further suggest a deeper origin, despite the relatively shallow water.

Figure 16. CH_4 concentration mapping in the air above Lake Orsa on June 27, 2024. Two distinct 10×10 m emission hotspots were identified, along with several smaller sources. In these areas, ebullition and diffuse emissions from the water surface were measured, and water samples were collected for isotopic analysis.

4.3 Floating chamber measurements

The floating chamber method was used to quantify the fluxes of CH_4 and CO_2 from water surfaces to the atmosphere (Figure 4). Measurements were performed both at background locations—i.e., outside identified hotspot areas—and in the vicinity of known ebullition sources, while deliberately excluding direct ebullition events. As described in Section 2.2, this method involves placing a chamber that floats on the water surface, isolating a portion of the air–water interface. The chamber captures gas exchange between the water and atmosphere, and fluxes are calculated by monitoring the change in gas concentrations inside the chamber over time. Measurements were conducted at multiple locations (Figure 17 and Table 2) on Lake Siljan (sampling points #1–7) and Lake Orsa (#8–16) (Figure 18). Measurements #8–16 are considered the most reliable due to successive improvements in the experimental setup. Sampling points #1, #9, and #10 serve as background references.

An example from Lake Orsa is shown in Figure 19, illustrating CH_4 and CO_2 concentration trends inside the floating chamber at sampling positions #11, #12, and #13. A sharp increase in concentration observed at 14:04 is interpreted as the entry of a gas bubble into the chamber, indicating an ebullition event.

Figure 20 presents the CH_4 fluxes calculated from the water surface at the various locations. As noted, all values represent diffuse emissions, since ebullition events such as the one in Figure 19 were excluded from the analysis.

The measured values are consistent with a recent study of Swedish lakes (8), which reported typical diffuse CH_4 emissions ranging from 0.02 to 0.05 $\mu g \cdot m^{-2} \cdot s^{-1}$, with nighttime emissions approximately 50% lower.

The data have been categorized into three emission classes:

- Background (< 0.02 $\mu g \cdot m^{-2} \cdot s^{-1}$): sampling points #5, #8, #9, #10, and #15
- Intermediate (0.02–0.05 $\mu g \cdot m^{-2} \cdot s^{-1}$): sampling points #1, #2, #3, #11, #12, #14
- High (> 0.05 $\mu g \cdot m^{-2} \cdot s^{-1}$): sampling points #4, #6, #7, and #13

Three of the four highest-emitting points (#4, #6, and #13) were located near known ebullition hotspots. The fourth, point #7, is believed to represent a more classical diffuse ebullition source, since it was not localized but spread over a larger shallow area with reeds and more organic sediment.

In Table 2, the measured area emissions by the floating chamber have been converted to estimated hourly emissions by assuming an active emission area. For the localized hotspots (sampling points #4, #6, and #13), an active area of 30 × 30 m was assumed. For the non-localized emission source at Ickviken (#7), a larger area of 100 × 100 m was used.

The sampling point #2 at Sollerön was initially selected as a background site based on earlier observations, but it showed higher-than-expected emissions, possibly due to influence from a nearby localized source. Ice wakes were frequently observed in this area as well. The remaining seven sampling points were located near known ebullition hotspots, with the exception of Sollerön (#1 and #2), where only limited ebullition was detected.

It should be noted that the sample size in this study is relatively small and that the floating chamber method has inherent limitations in capturing spatial heterogeneity. A larger number of measurements is typically required to fully account for variability. Nevertheless, a clear trend was observed: sites with active or nearby ebullition consistently exhibited elevated diffuse CH_4 emissions—up to four times higher than those reported from other Swedish lakes.

Figure 17. Overview of all performed Floating chamber measurements, shown with yellow markers.

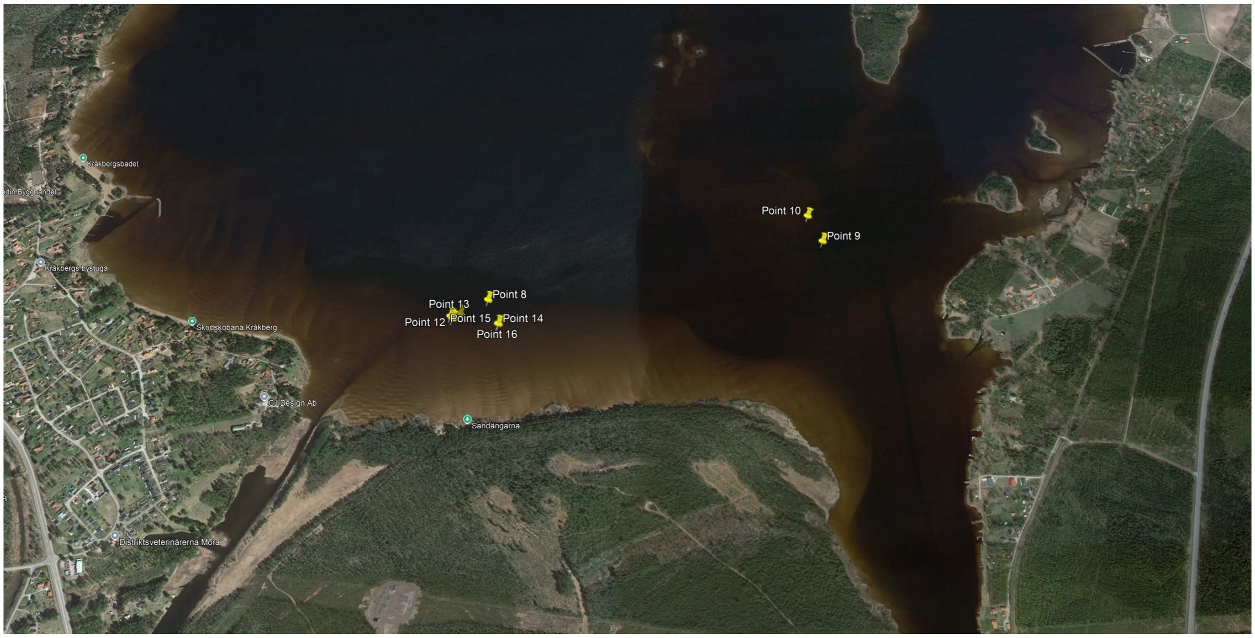

Figure 18. Floating chamber measurements on June 27 at the southern shore of Lake Orsa, shown with yellow markers.

Table 1 Floating chamber measurements performed during the 2024 campaign.

Sampling location	Time	Longitude	Latitude	Depth	Comment
1	2024-06-25 12:58:23	14.6104675	60.95499	~6 m	NE Rattholmen near Sollerön in Lake Siljan
2	2024-06-25 14:45:00	14.5987175	60.94644	~6m	SW Rattholmen near Sollerön in Lake Siljan
3	2024-06-25 16:42:24	14.60438978	60.96382253	2-4 m	Fudalsviken at Lake Siljan, outside entrance to harbor.
4	2024-06-25 16:49:13	14.60366219	60.96718127	2-4 m	Fudalsviken at Lake Siljan, outside entrance to harbor.
5	2024-06-26 09:55:00	14.80995508	60.89186503	>50 m	Lake Siljan, 1.6 km S Kroksud,
6	2024-06-26 12:10:00	15.0419104	60.90811746	< 2 m	Åkernäsviken near Vikarbyn, in Lake Siljan
7	2024-06-26 15:06:13	14.96526304	60.90127724	~1 m	Ickviken in Lake Siljan
8	2024-06-27 08:29:00	14.55279878	61.03200094	5-12 m	SW Lake Orsa, source 1
9	2024-06-27 09:18:00	14.57043585	61.03391852	~12 m	SE Lake Orsa, background
10	2024-06-27 09:25:00	14.56953195	61.03477141	~12 m	SE Lake Orsa, background
11	2024-06-27 13:51:00	14.55145487	61.03156781	2-5 m	SW Lake Orsa, source #2
12	2024-06-27 14:02:00	14.55109144	61.03140936	2-5 m	SW Lake Orsa, source #2
13	2024-06-27 14:08:30	14.55094419	61.03149202	5-12 m	SW Lake Orsa, source #2
14	2024-06-27 15:01:00	14.55356977	61.03124008	2-5 m	SW Lake Orsa, source #2
15	2024-06-27 15:05:00	14.55355022	61.03124116	2-5 m	SW Lake Orsa, source #2
16	2024-06-27 15:09:00	14.55357368	61.03124521	1-5 m	SW Lake Orsa, source #2

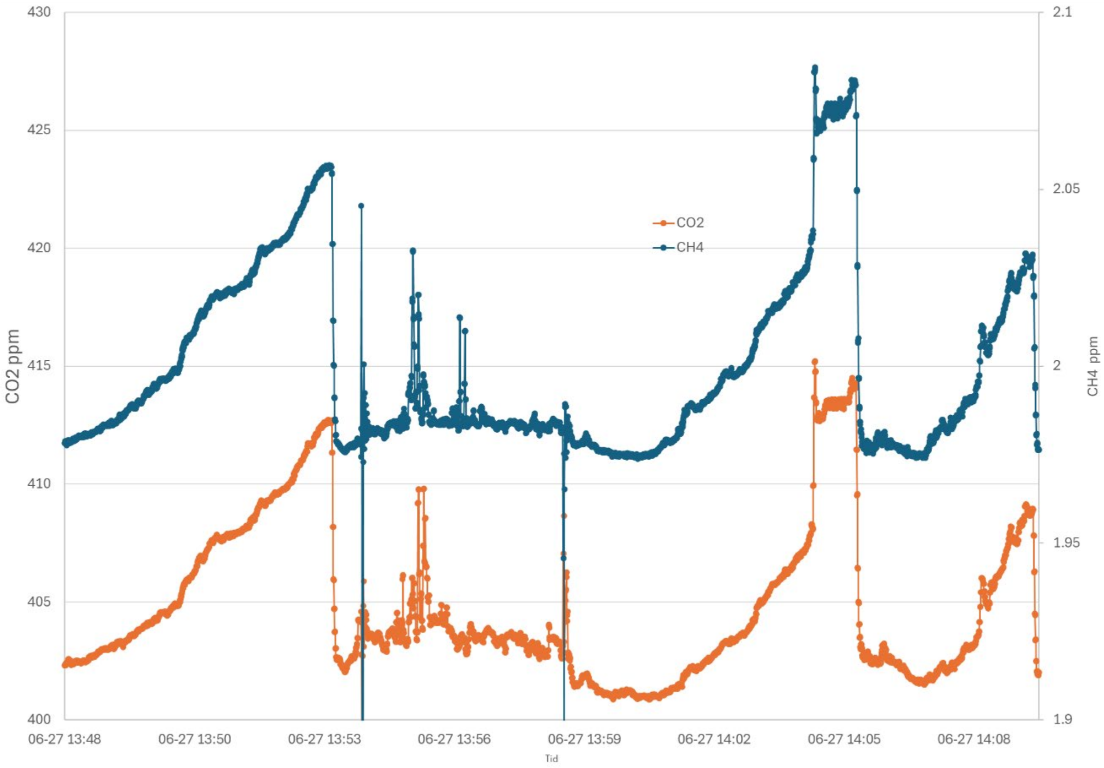

Figure 19. Example of concentration measurements inside a floating chamber on Lake Orsa, near the CH_4 hotspot at sampling positions #11, #12, and #13. Note the steep slope at 14:04, interpreted as the entry of a bubble into the floating chamber, corresponding to ebullition.

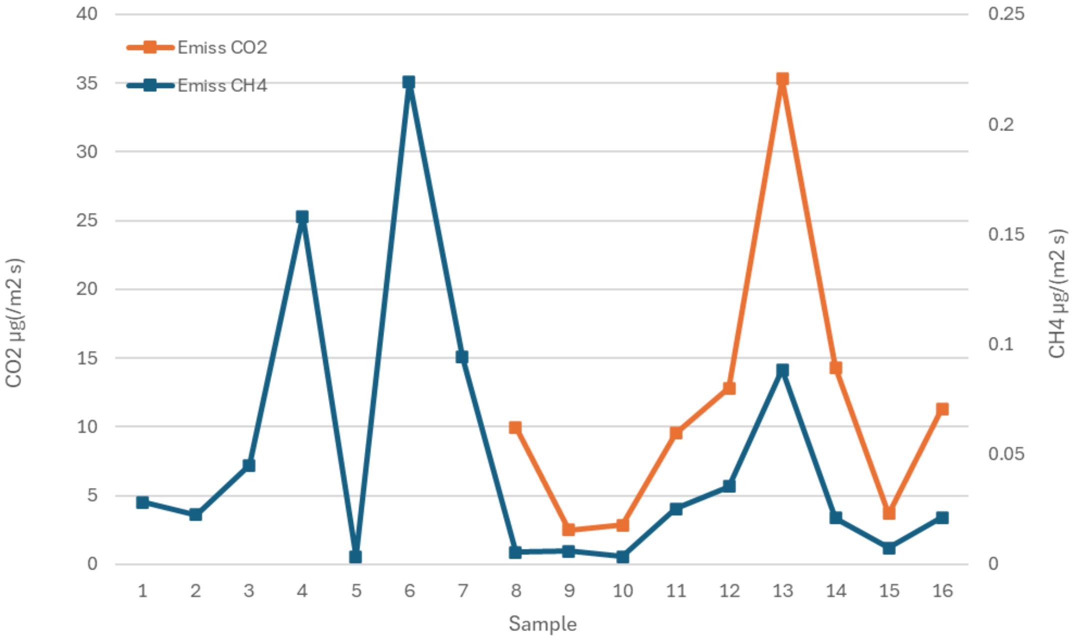

Figure 20. Emissions from the water surface during floating chamber measurements at various points on Siljan (#1–7) and Lake Orsa (#8–16). Measurements #8–16 are the most reliable due to the experimental setup. Measurements #1, #9, and #10 represent background levels.

Table 2. Summary of key sampling points associated with high methane (CH_4) emissions identified during the June 2024 campaign around Lake Siljan and Lake Orsa. The CH_4 flux using the floating chamber method (in $\mu g \cdot m^{-2} \cdot s^{-1}$), and the estimated diffuse emission rate from water surface (in g/h), assuming a representative active area are shown. The values exclude direct ebullition events and reflect diffuse surface emissions.

Sampling Point	Description / Location	Flux ($\mu g \cdot m^{-2} \cdot s^{-1}$)	Estimated diffuse Emission (g/h)
#4	Fudalsviken (Figure 12) Localized ebullition source, 30×30 m area	0.15	0.5
#6	Åkernäsviken, Vikarbyn, (Figure 14, Figure 15) Localized ebullition source, 30×30 m area	0.22	0.7
#7	Ickviken (Figure 13) Non-localized ebullition, 100×100 m area	0.095	3.5
#13	SW Lake Orsa (Figure 16) Localized ebullition source #2, 30×30 m area	0.088	0.3

4.4 Emission measurements by the Tracer dispersion method (TDM)

The Tracer Dispersion Method (TDM) was applied at four CH_4 concentration hotspots with identified ebullition sources. A small dinghy was positioned near each ebullition source, from which the trace gas ethane was released in a controlled manner at rates ranging from 0.5 to 4 kg/h. Measurements were carried out by repeatedly transecting the plume orthogonally—driving back and forth across the plume multiple times over a period of approximately 30 minutes. The integrated masses of both ethane and CH_4 were measured along the transects, summed over the full measurement period, and used to calculate CH_4 emission rates based on Equation 6 in Section 2.4.

Table 3 presents the results for all TDM measurements performed, including two sources in the southern part of Lake Orsa and two sources in Lake Siljan. Measurements were taken approximately 50–100 meters downwind using an Aeris and Picarro. The measurement data results are also shown in the following graphs.

In Figure 21, TDM measurements are shown for an active CH_4 ebullition source area (estimated as 30 × 30 m) at the port inlet of Fudalsviken in North Siljan. The ethane spikes are sharper than those for CH_4, indicating that the emission source was more widespread for CH_4. The source was persistent for at least an hour and was also observed four hours earlier during a mapping exercise.

Figure 22 displays TDM measurements for the ebullition source at the old steamboat pier at Vikarbyn, Lake Siljan. Measurements were taken approximately 50 meters downwind of the source using an Aeris and Picarro, as shown in Figure 8. CH_4 is considerably more widespread than ethane, indicating a larger source area, likely around 30 × 30 m, compared to the single release point of ethane. The difference between CH_4 and ethane appears larger than what it actually is, due to the relatively short transects and the slow movement of the boat.

Figure 23 and Figure 24 show TDM measurements at two nearby ebullition sources at Lake Orsa, separated by about 100 m. These sources were also observed during the first campaign in 2023.

The ebullition reported in Table 3 can be compared to daytime ebullition rates of 0.05 $\mu g \cdot m^{-2} \cdot s^{-1}$ measured at other Swedish lakes (8). For an emission area of 30 × 30 m, such a flux would correspond to approximately 0.16 g/h. This reference value is:

- 200–300 times lower than the emission rate observed at Lake Orsa
- ~2000 times lower than the rate measured at the steamboat pier in Vikarbyn, and
- ~130 times lower than the rate measured at Fudalsviken.

In conclusion, the emissions at the four hotspots listed in Table 3 are predominantly due to ebullition rather than diffuse emissions, sd supported by the floating chamber results in Table 2. Moreover, these emissions are significantly stronger—by two to three orders of magnitude—than typical lake ebullition rates, indicating that the studied sources are unusually strong and likely unique. It should be noted that only a limited number of measurements transects were conducted, and additional data are needed to improve the statistical robustness of the emission estimates. Nonetheless, the concentrations measured near the hotspots in Lake Orsa were similar during both campaigns, suggesting that the ebullition rates were of the same order of magnitude across time.

Table 3. Methane leakage from ebullition sources identified at concentration hotspots in Siljan and Lake Orsa. Results from trace gas releases of ethane (0.5–4 kg/h) at active emission sources and downwind measurements 50–200 m away using Picarro (CH_4) and Aeris (C_2H_4) laser sensors. Typical ebullition on other Swedish lakes is also shown, assuming a 30 by 30 m release area.

Date time	Position	Source Longitude	Source Latitude	Tracer release kg/h	# Transects	CH_4 g/h this study	CH_4 g/h Literature (8)
25/6-2024 17:09-17:51	Fudalsviken, Lake Siljan	14.60438978	60.96382253	1.52	8	21	0.16
26/6-2024 12:59-13:26	Vikarbyn, Lake Siljan	15.0419104	60.90811746	4.76	4	309	0.16
27/6-2024 10:40-11:36	SW Lake Orsa, source 1	14.57043585	61.03391852	1.58	4	33	0.16
27/6 -2024 14:35-14:55	SW Lake Orsa, source 2	14.55094419	61.03149202	0.43	4	43	0.16
SUM						406	0.64

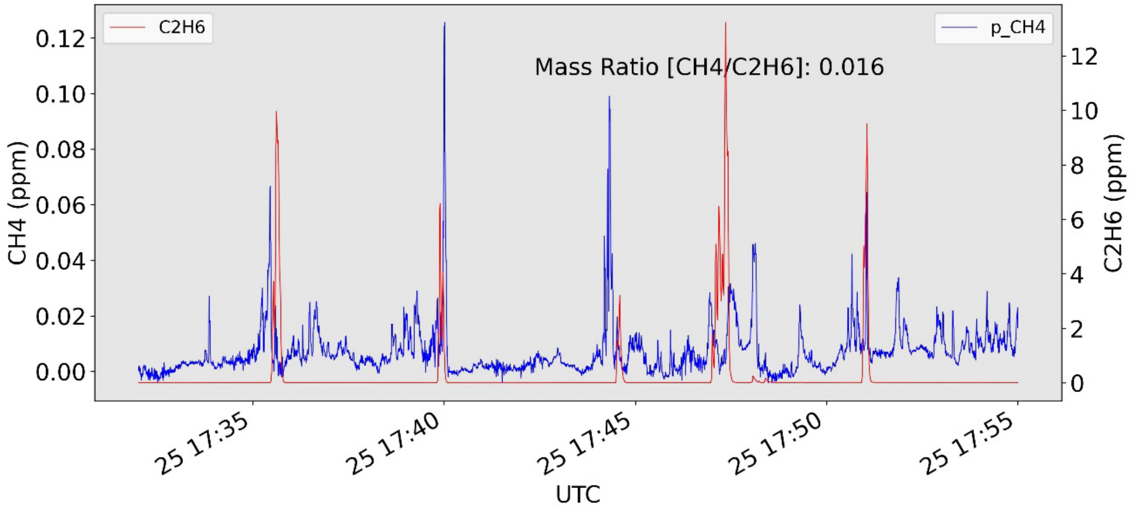

Figure 21. TDM measurements using ethane as tracer over an active CH_4 leakage area (30 × 30 m^2) at a ebullition source at port inlet Fudalsviken, N Siljan . Measurements were taken approximately 50-100 meters downwind using an Aeris and Picarro. The CH_4/C_2H_6 mass ratio is 1.6%.

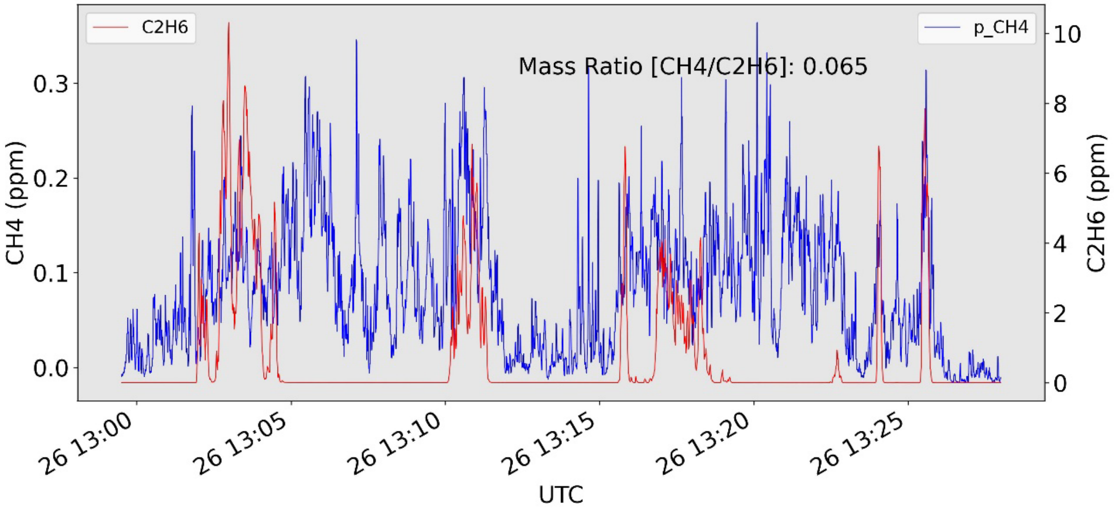

Figure 22. TDM measurements using ethane as tracer over an active CH_4 leakage area at a ebullition source at old steamboat pier at Vikarbyn lake Siljan. Measurements were taken approximately 50 meters downwind using an Aeris and Picarro, as shown in Figure 6. The CH_4/C_2H_6 mass ratio is 6.5%.

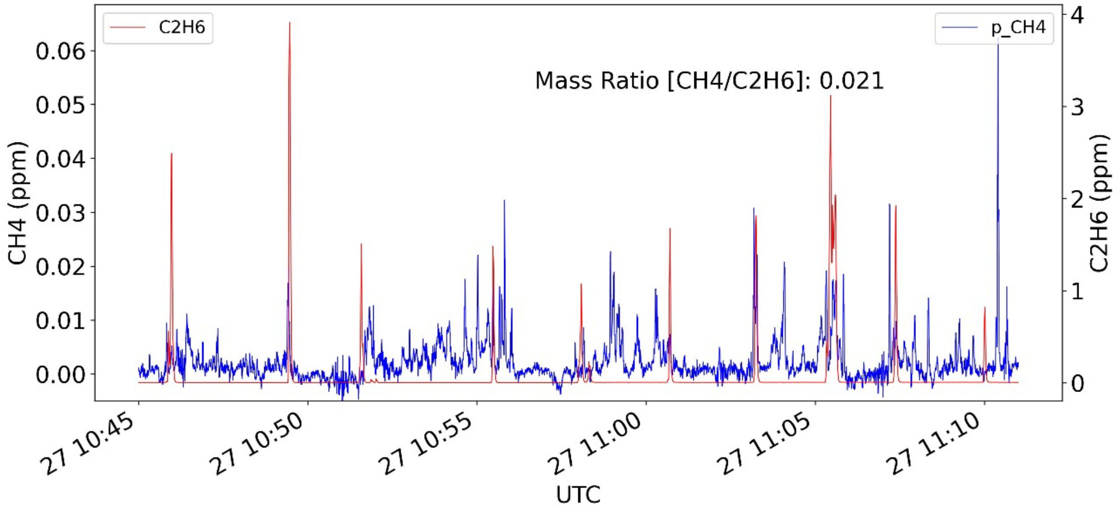

Figure 23. TDM measurements using ethane as tracer over an active CH_4 leakage area (30 × 30 m^2) at a source 1 at Lake Orsa. Measurements were taken approximately 50–200 meters downwind using an Aeris and Picarro. The CH_4/C_2H_6 mass ratio is 2.1%.

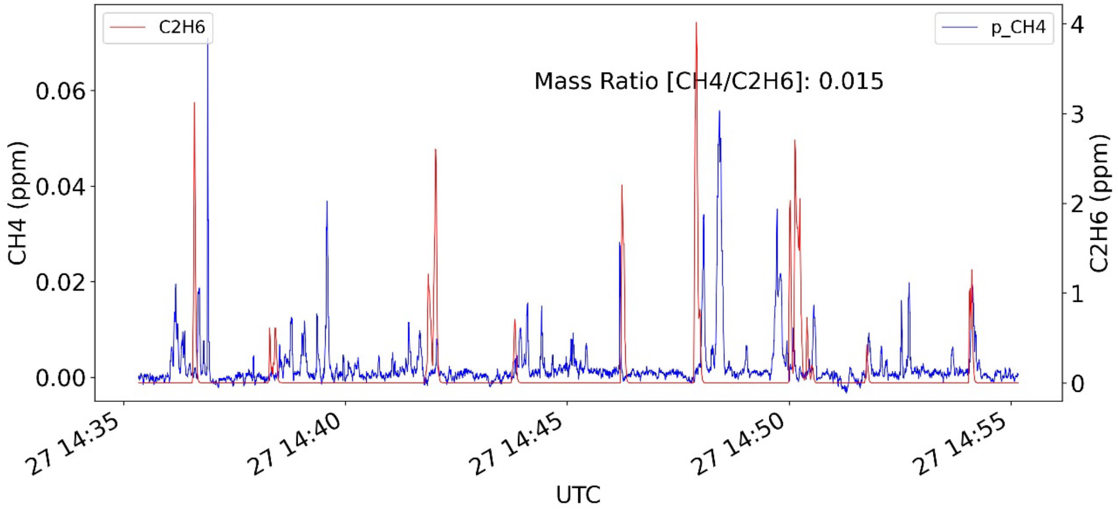

Figure 24. TDM measurements using ethane as tracer over an active CH4 leakage area (30 × 30 m^2) at a source 2 at Lake Orsa. Measurements were taken approximately 50–200 meters downwind using an Aeris and Picarro The CH_4/C_2H_6 mass ratio is 1.5%.

4.5 Isotope Analysis and Ethane/CH_4 Ratio.

In this study, ambient background values of CH_4 were measured at the laboratory in Gothenburg, corresponding to a $\delta^{13}C$ value of -52‰. In contrast, thermogenic CH_4, formed through the thermal breakdown of organic material, has less negative $\delta^{13}C$ values (around -40‰). CO_2 produced from the oxidation of biogenic CH_4 exhibits higher $\delta^{13}C$ values, ranging from -30‰ to +10‰, depending on the degree of fractionation during oxidation.

In Campaign 1, CH_4 and ethane measurements were taken from a test borehole near the Mora field, showing an ethane-to-CH_4 ratio of 1%, consistent with other analyses (22) and indicating a weak thermogenic origin. As described in the introduction, the same study (22) showed a very negative $\delta^{13}C$ CH_4 value and slightly negative to positive $\delta^{13}C$ CO_2 values. It was concluded that this corresponds primarily to microbial CH_4 (Table 3).

During the measurement campaign, a raw water sample was collected from Lake Orsa from ebullition source 2; this sample also contained sand. Another bottle sample was collected in February from Lake Siljan and contained only water. The results are shown in Table 2. The sample from Lake Orsa stands out due to its very high concentrations of both CH_4 and CO_2 relative to the background. The $\delta^{13}C$ value of CH_4 was -42‰, indicating a thermogenic origin. This contradicts the abovementioned analysis of borehole measurements (22), which show much lower values and are primarily of bacterial origin. However, it was not part of the project plan to collect the aforementioned samples for isotope analysis, and the sampling process introduced many potential sources of error, making the overall measurements uncertain and difficult to interpret. These values are retained for comparison with future samples but not used in the conclusions of this study. In the lake measurements, the gas concentrations were insufficient to detect the ratio of ethane to CH_4 at the 1% level, as measured at the borehole. Consequently, the results are inconclusive regarding the origin of gas at the ebullition sources based on this analysis.

Table 4. Isotopic analysis of $\delta^{13}C$ in CH_4 and CO_2 in the headspace of water samples from June 2024 was conducted using laboratory analysis with Picarro G2131 from Göteborg University (Santos and Yau). The background concentration for CH_4 and CO_2 was approximately 1.95 ppm and 436 ppm, with $\delta^{13}C$ values of -52‰ and -9‰ for CH_4 and CO_2, respectively.

Site		CH_4 ppm	$\delta^{13}CH_4$	CO_2 ppm	$\delta^{13}CO2$	$\Delta CO_2/\Delta CH_4$	$\Delta C_2H_6/\Delta CH_4$
Sollerön, Lake Siljan (background)	March 2024	2.02	-51‰	1003	-16.45‰	8100	NA
Orsasjön, (Ebullition source)	June 2204	32	-42‰	12132	-19‰	387	NA
Boreholes from (22) and this study			-57‰ to -63‰		-14‰ to +6‰		1%

5. Discussion

CH_4 measurements conducted at lakes surrounding the Siljan Ring revealed distinct hotspots where CH_4 is consistently leaking, forming detectable concentration plumes in the air through ebullition. The lakebeds in the Siljan region consist predominantly of sand, and the lakes Siljan and Orsa are generally deep with well-circulating water. All identified leaks were located in shallow areas (2–5 meters deep) under non-stagnant conditions. These emissions were highly localized, making them atypical for diffuse CH_4 release commonly observed in lakes.
Mapping identified active ebullition sources in Lake Orsa (2–4 locations), Fudalsviken (1 location), and Vikarbyn (at the old steamship pier and Åkernäsviken). Elevated emissions were also observed in both air and water at Ickviken, where shallow water and sediment presence contributed to classical lake sediment emission activity. Floating chamber measurements showed CH_4 fluxes ranging from 0.05 to 0.2 $\mu g\ m^{-2}\ s^{-1}$, with CO_2 fluxes approximately 100 times higher. The floating chamber CH_4 emission values exceed typical background lake emissions, which usually range from 0.02 to 0.05 $\mu g\ m^{-2}\ s^{-1}$ showing that the emissions at the hotspots are usually high. The TDM measurements showed that ebullition sources had emissions ranging from 20 to 300 g/h—several orders of magnitude higher than the diffuse fluxes measured by floating chambers—highlighting the dominant role of ebullition at these sites. Moreover, these emissions are significantly stronger—by two to three orders of magnitude—than typical ebullition rates observed in lakes, indicating that the studied sources are exceptionally strong and likely unique.
Total CH_4 emissions from the identified hotspots were estimated at approximately 3.5 tons per year (equivalent to 85 tons CO_2). In addition, diffuse emissions near these hotspots were estimated at 50 kg per year (equivalent to 1 ton CO_2). Combined, these emissions represent a small fraction—around 20 parts per million—of Sweden's national CH_4 emissions, according to the 2023 National Inventory Report (21). However, it is likely that many more ebullition hotspots exist. This study focused on the eastern shores of Lake Siljan and the southern part of Lake Orsa, but additional sources may be present, particularly at greater depths where detection is more challenging.
Given the limited number of measurements and potential artifacts from sampling procedures, the origin of the gas—whether thermogenic or biogenic—remains uncertain. Further studies are required to analyze both the volatile organic compound (VOC) content and the $\delta^{13}C$ isotopic composition to determine the source more precisely.
The CH_4 sources identified in the Siljan Ring are unique due to their persistence, strong localization, and temporal variability—traits that make them difficult to detect using conventional techniques.
Traditional methods such as floating chambers and eddy correlation often fall short when it comes to capturing this type of localized emissions since they are designed to measured area emissions. Continued research is essential to assess the temporal dynamics of these emissions, identify their origins, and expand source mapping to other parts of the region. Depending on future findings, these results could have implications for emission estimates from other lake systems, contribute to refining Sweden's natural climate budget, and support the development of mitigation strategies to manage such emissions effectively.

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7. Acknowledgement

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