

LETTER

Tracing the origins of metalimnetic methane in a stratified large lake: Insights from high-resolution mapping and three-dimensional hydrodynamic modeling

Santona Khatun ^{1,2*} Roberto Grilli ³ Abolfazl Irani Rahaghi ^{4,5} Tomy Doda ²
Damien Bouffard ^{2,6} Carsten J. Schubert ^{6,7} Jérôme Chappellaz ¹ Marie-Elodie Perga ²

¹School of Architecture, Civil and Environmental Engineering, Environmental Engineering Institute, Smart Environmental Sensing in Extreme Environments (SENSE), École Polytechnique Fédérale de Lausanne (EPFL Valais Wallis), Sion, Switzerland; ²Faculty of Geosciences and Environment, Institute of Earth Surface Dynamics, University of Lausanne, Lausanne, Switzerland; ³University of Grenoble Alpes, IRD, CNRS, INRAE, Grenoble INP, IGE, Saint-Martin-d'Hères, France; ⁴Department of Surface Waters—Research and Management, Eawag, Swiss Federal Institute of Aquatic Science and Technology, Dübendorf, Switzerland; ⁵Department of Geography, University of Zurich, Zürich, Switzerland; ⁶Department of Surface Waters—Research and Management, Eawag, Swiss Federal Institute of Aquatic Science and Technology, Kastanienbaum, Switzerland; ⁷Department of Environmental Systems Science, ETHZ, Zürich, Switzerland

Scientific Significance Statement

Methane emissions from lakes are an important component of the global carbon cycle, yet the processes driving methane accumulation in oxygenated layers, the so-called methane paradox, remain unclear. This study shows that in Lake Geneva, methane-rich water from the Rhône River delta spreads across the metalimnion mainly through currents along density layers and circular circulation patterns (gyres). By associating 3D hydrodynamic modeling with spatially distributed methane measurements, our work offers a mechanistic resolution of the methane paradox and underscores the broader role of river-lake connectivity in regulating greenhouse gas dynamics in freshwater ecosystems.

Abstract

Methane (CH₄) accumulation in the oxic metalimnion is a common yet poorly resolved feature of deep stratified lakes. In Lake Geneva, lateral transport from the Rhône delta has been suggested but lacks spatial validation. Here, we quantify the role of lateral advection in shaping metalimnetic CH₄ using high-resolution in situ CH₄ measurements, δ¹³C-CH₄ analyses, and a three-dimensional Lagrangian particle-tracking model. Elevated metalimnetic CH₄ (up to 1143 nM) near the Rhône delta becomes isotopically enriched, likely from prolonged residence and partial oxidation during summer stratification. While additional sources may contribute, comparisons of in situ measurements with particle-tracking simulations suggest circulation gyres may distribute deltaic CH₄. Moreover, isotope- and transport-based indicators suggest more coherent coupling in the epilimnion than

*Correspondence: santona.khatun@epfl.ch; santona.khatun@unil.ch

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in the metalimnion. These results demonstrate that horizontal advection can be an important driver of metalimnetic CH₄ and highlight the importance of circulation-driven lateral transport for assessing CH₄ fluxes from large, stratified lakes.

Lakes are increasingly recognized as significant methane (CH₄) emitters, prompting improved global emission estimates (Rosentreter et al. 2021; Johnson et al. 2022; Saunio et al. 2024) and raising questions about their response to environmental changes (Beaulieu et al. 2016; Sepulveda-Jauregui et al. 2018). Understanding CH₄ dynamics in lakes is challenging due to substantial spatial and temporal variability in concentrations, particularly in large systems (Loken et al. 2019; Peacock et al. 2023). This heterogeneity reflects various CH₄ sources, including groundwater inflow, anaerobic methanogenesis (Bastviken et al. 2008, 2011), and oxic methane production (OMP, Grossart et al. 2011; Donis et al. 2017; Khatun et al. 2019). Moreover, CH₄ distribution variability is influenced by the rates and ranges of transport processes such as diffusion, ebullition, and advection (Tang et al. 2016; Peeters et al. 2019; Liu et al. 2024). These dynamics can bias CH₄ budgets when extrapolated from single-point measurements (Loken et al. 2019). Intertwined production, consumption, and transport processes also lead to confounding effects when quantifying the dominant CH₄ sources in lakes. Metalimnetic CH₄ peaks in stratified lakes have been attributed variously to OMP (Günthel et al. 2019; Hartmann et al. 2020), lateral input from sublittoral zones (Encinas Fernández et al. 2016; Peeters and Hofmann 2021), or limited oxidation (Donis et al. 2017; Liu et al. 2024). Thus, advancing our understanding of methane fluxes in lakes necessitates the adoption of a three-dimensional (3D) perspective (Guo et al. 2023).

Early one-dimensional (1D) lake models have focused primarily on sediment-atmosphere CH₄ fluxes via diffusion and ebullition but lacked spatial resolution, neglected lateral transport components, or assumed it to be diffusive only (Tan and Zhuang 2015; Stepanenko et al. 2016; Encinas Fernández et al. 2016; Schmid et al. 2017; DelSontro et al. 2018). Recent studies highlight the dominant role of advection, far faster than diffusion, in horizontal CH₄ distribution (Peeters et al. 2019; Doda et al. 2024) and the influence of lake morphology on ebullition variability (Sø et al. 2025). However, most models still simplify CH₄ budgets to 1D simulations using only vertical fluxes, even when a 3D lateral transport model exists (Guo et al. 2023). Recent hydrodynamic modeling approaches using Lagrangian particle tracking (Cimatoribus et al. 2018, 2019; Baracchini et al. 2020) offer promising tools for simulating lateral CH₄ transport. In addition, measurement-based approaches to CH₄ mapping remain labor-intensive when relying on discrete water sampling and off-line analysis. Automated surface mapping tools like FLAME can resolve 2D patterns (Loken et al. 2019), while newly available submersible sensors enable vertical profiling, including isotopic analysis (Grilli et al. 2023).

In Lake Geneva, earlier studies based on discrete sampling (Khatun et al. 2024) suggested that lateral transport redistributes CH₄ from sublittoral zones near the Rhône delta across tens of kilometers in the metalimnion. This work proposed that advective CH₄ transport from the Rhône delta, rather than OMP, was the dominant source of metalimnetic CH₄. However, the previous dataset lacked sufficient spatial resolution and hydrodynamic validation, limiting the ability to trace CH₄ source connectivity and transport mechanisms across the 580 km² lake basin.

To address these gaps, we conducted a combined observational and modeling study focused on the role of lateral advection in shaping CH₄ distributions in Lake Geneva. Our hypothesis does not include OMP, instead targeting CH₄ produced near the Rhône delta and redistributed by lateral advection. We combined high-resolution in situ CH₄ measurements from a SubOcean probe (membrane inlet-laser spectrometer) with lab-based isotopic ($\delta^{13}\text{C-CH}_4$) analyses and a 3D Lagrangian particle tracking model. The isotopic data allowed us to trace CH₄ sources and oxidation, while the 3D particle model explicitly simulates lateral flows across the lake. By comparing observed CH₄ distributions in the epilimnion and metalimnion with particle trajectories from hypothesized source zones, we quantify the role of lateral transport and better constrain CH₄ distribution in a large-stratified lake. Together, these datasets reveal how lateral advection governs metalimnetic CH₄ patterns by connecting deltaic CH₄ sources to offshore locations across Lake Geneva.

Methodology

Site description

Lake Geneva, a large deep monomictic lake on the France-Switzerland border (Fig. 1), is the largest lake of Western Europe. Due to the unique hydrodynamics and morphology of Lake Geneva, metalimnetic CH₄ has previously been hypothesized to be advected from the Rhône River's deltaic hotspot (Khatun et al. 2024). The lake's outlet is located in the western part, flowing into France through the Geneva canton.

Real-time methane measurement

An extensive survey from the Rhône River delta toward the LÉXPLORE floating platform (Fig. 1) was conducted using the SubOcean probe (Grilli et al. 2018). It is a high-sensitivity and fast-response in situ membrane inlet-laser spectrometer instrument (Response time, $t_{90} \sim 30$ s, detection limit 1 nM, and concentration span up to 15 μM). We deployed the probe onboard the FOREL research vessel “La Licorne” on June 28 and 29, 2023. For real-time dissolved CH₄ concentrations,

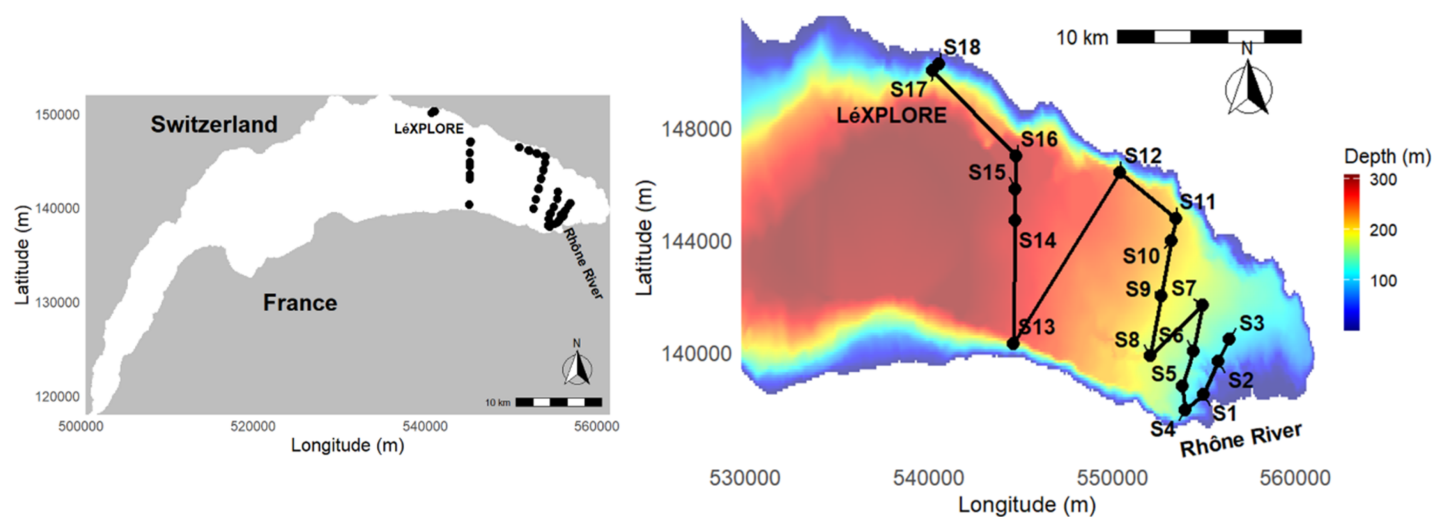


Fig. 1. Bathymetry and distribution of sampling sites in Lake Geneva. Real-time methane distribution across the lakes was obtained using the SubOcean probe for different transects and stations. We named the 18 sampling stations (S1–S18) along the transects to compare the profiles with discrete water samplings. Water samples were collected for dissolved CH_4 concentration and isotopic analysis at 11 stations (e.g., S2, S3, S5–S10, S12, S15, S17). Station S1 is close to the Rhône River mouth, and station S18 is in the vicinity of the LEXPLORE research platform.

the probe was deployed at multiple stations and ensured continuous measurements along the four transects (Fig. 1). For some of the sampling stations, full-depth water-column CH_4 profiles were acquired, although we only focus on the top 27 m in this study. These in situ measurements were validated against laboratory measurements carried out on discrete samples, collected at 11 stations during the same days but with a different boat. The maximum time span between continuous in situ measurements and discrete sampling was 1 h. Water sample collection and analysis details are provided in the Supporting Information.

3D hydrodynamics model and Lagrangian particle tracking

The 3D particle tracking model assesses the movement of water parcels within the lake, accounting only for advective transport (i.e., neglecting diffusion from random particle displacement). The MIT General Circulation Model (Marshall et al. 1997) was employed to simulate the lake hydrodynamics and potential transport of dissolved CH_4 concentration over the sampling days. The model configuration follows Irani Rahaghi et al. (2024), with the Rhône inflow excluded, as the focus here is on basin-scale transport processes. The model was initialized (i.e., from the zero-velocity field) with the horizontally homogeneous temperature condition on 31 May 2023. Reanalysis of meteorological data over the lake from the COSMO-1 numerical weather model provided by MeteoSwiss (<https://www.meteoschweiz.admin.ch/>) on a $1.1 \text{ km} \times 1.1 \text{ km}$ grid was used as model forcing. The lake hydrodynamics were then simulated until 29 June 2023, but only the results during 24–29 June were used for further analysis, acknowledging the model spin-up time of ~ 3 weeks (Irani Rahaghi et al. 2024).

From now on, we will use the word “particle” to refer to the tracked water parcels in the numerical model. We focused on the lake transport, not on the direct intrusion of the Rhône River into the metalimnion. Particles were continuously released between 24 and 29 June near the Rhône River delta at 7–22 m (see Supporting Information), corresponding to the location of deltaic CH_4 sources (Khatun et al. 2024). For each lake grid cell in the epi- and meta-limnion, we quantified the probability of particle presence as the fraction of the measurement period (28 June 11:30–29 June 17:30). More details on model setup, water-column variability, and validation can be found in Safin et al. (2022), Irani Rahaghi et al. (2024), and in the Supporting Information (see Supporting Information and Figs. S1–S3).

Oxidation-derived and model-derived fractionation

We applied a Rayleigh fractionation model to calculate the fraction of methane remaining, $f_{\text{ox_iso}}$, to estimate the time scale over which methane undergoes oxidation. Then, it is used to estimate the isotope-derived oxidation time ($t_{\text{ox_iso}}$) assuming first-order decay. Moreover, transport-derived fractionation, $f_{\text{ox_model}}$, for the model-based particles were additionally estimated using an exponential decay formulation based on particle age ($t_{\text{ox_model}}$, see Supporting Information).

Results

Spatial methane dynamics in Lake Geneva

The lake was fully stratified with a metalimnion at 7–22 m depth, separating cold waters from warm surface (approximately 22.4°C , Supporting Information Figs. S3 and S4). Dissolved oxygen (DO) remained high ($> 8 \text{ mg/L}$, i.e., at $\sim 90\%$

saturation), peaking near the thermocline coinciding with maximum turbidity (Supporting Information Figs. S3 and S4).

Depth-averaged epilimnetic (0–6 m) CH₄ concentrations ranged from 36 to 162 nM, with lowest values near the Rhône delta (S1) and highest near LÉXPLORE platform (S18), respectively (Fig. 2A). Local (non-depth-averaged) CH₄ concentrations varied from 13 nM at 0.5 m (S16) to 277 nM at 1.5 m (S18), respectively (Supporting Information Fig. S5). High concentration of 108 nM was detected at 0.5 m between stations S1 and S2, while 106 nM was observed at 0.5 m at S18. Most stations showed average epilimnetic CH₄ concentration > 41 nM, with some stations showing increasing CH₄ starting from 2 m depths (e.g., S2–S3, S8–S9, S15–S16, and S17), while others at deeper depths (> 6 m).

As opposed to a 1D system such as riverine water with comparatively simpler transport, CH₄ concentrations in the metalimnion (6.1–20 m) varied spatially, and were not simply

proportional to distance from shore or river mouth. Metalimnetic depth-averaged CH₄ concentrations ranged from 79 to 353 nM, while local values reached up to 1143 nM between S1 and S2 at 11 m (Fig. 2B,C; Supporting Information Fig. S6). Elevated metalimnetic CH₄ concentrations were also observed at southern nearshore sites S8 and S9 (Fig. 2C). Moreover, S18 showed a distinct peak of 438 nM at 14 m, suggesting an additional CH₄ source around this near-shore location. Below 20 m (hypolimnion), CH₄ typically declined, except near the Rhône delta (Fig. 2C). Notably, peaks > 1000 nM and 454 nM at 21.5 m and 45 m depths, respectively, were observed between S1–S2 and S1–S4.

Insights into the physical transportation of deltaic methane

The resulting particle trajectories offer a statistical perspective on the dominant water pathways, enabling us to trace

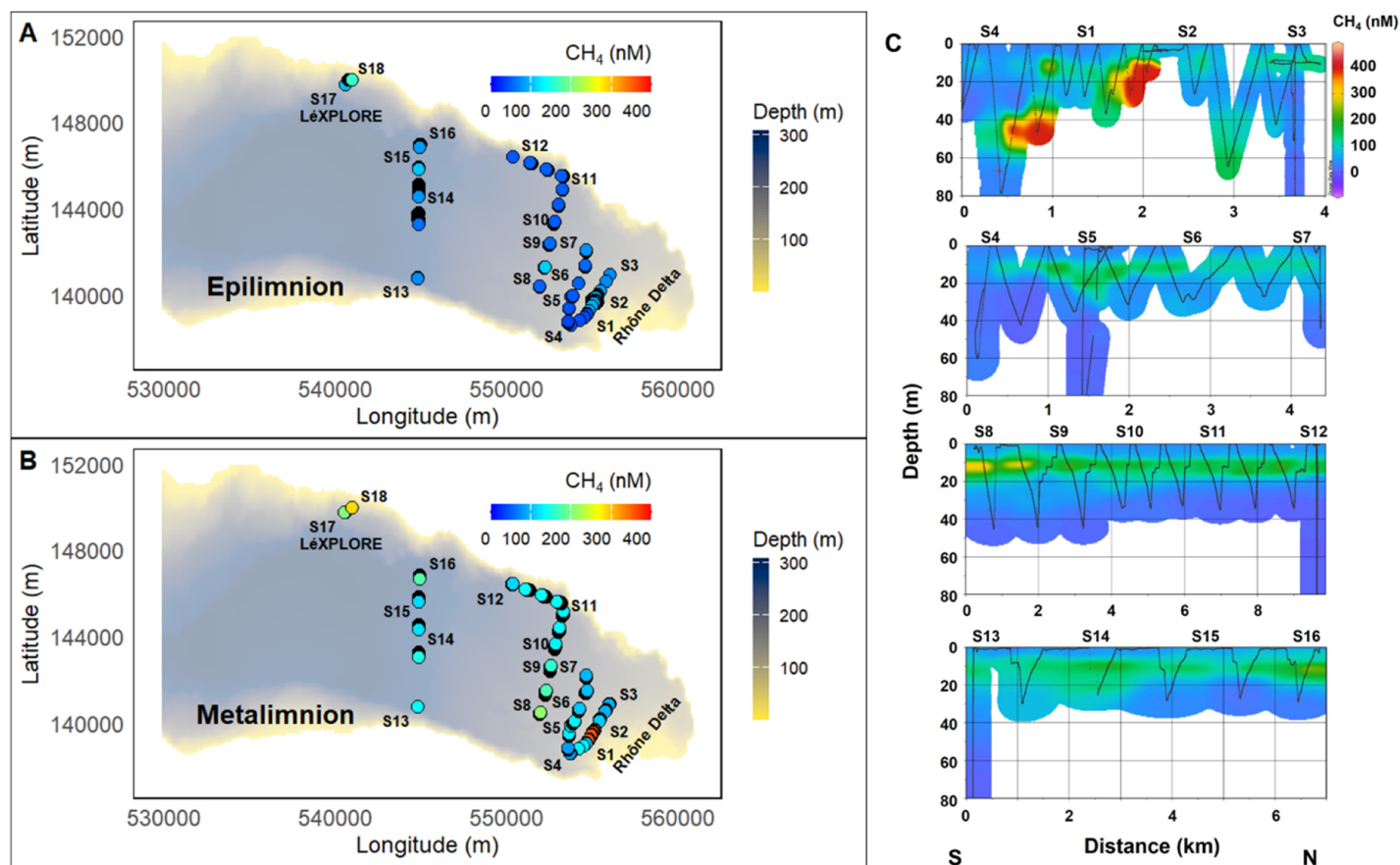


Fig. 2. Spatial methane mapping from the river delta toward the central lake. (A) and (B) show the depth-averaged distribution of epilimnetic (0–6 m) and metalimnetic (6.1–20 m) CH₄ concentrations from SubOcean data. The blue-red color scale represents the spatial variations of the depth-averaged CH₄ concentration (nM) for 18 stations in both layers. Values exceeding 400 nM are clipped at this upper limit to prevent distortion of the color gradient and to enhance spatial comparability. Bathymetry is shown using a yellow-bluish color scale (light yellow for shallow areas; dark blue for deeper zones). (C) Continuous metalimnetic CH₄ mapping along four transects. The colored scale represents the raw CH₄ values clipped at 400 nM. The black lines indicate sampling depth spans with spatial coverage among stations using the SubOcean probe. The spatial extrapolation around each black line is conducted using weighted-average gridding with automatically determined scale length. A transect distance from the southern shore (S) station to the northern shore (N) station is visualized as a 2D section (depth vs. distance) using Ocean Data View software (ODV 5.8.2, <https://odv.awi.de>).

the advective transport of the deltaic CH_4 produced near the river mouth. The results showed complex but distinguishable transport patterns (Supporting Information Figs. S1 and S2), including northward transport in both epilimnion and metalimnion, consistent with Coriolis effects (Fig. 3A,B). In our numerical setup, most particles were released in the metalimnion. Still, given the vertical variability of the thermocline depth combined with internal waves and basin-scale gyres, more than 3 d of particle release (Supporting Information Fig. S1, the full model simulation), a smaller fraction of the particles was directly released in the epilimnion, allowing transport patterns to be resolved in both layers. A basin-scale counterclockwise gyre was evident in both layers, with fewer particles in its center. This low-transport zone aligns with areas of reduced CH_4 concentrations near S14 (Fig. 2A), suggesting physical isolation (i.e., an upwelling region) may limit deltaic CH_4 accumulation there.

Measured CH_4 and modeled particle distributions are compared across stations to assess hydrodynamic controls on spatial CH_4 patterns, assuming a single source. Correlations are

moderate in the epilimnion ($r^2 = 0.41$, $n = 18$, $p < 0.001$; Fig. 3C) and slightly stronger in the metalimnion ($r^2 = 0.48$, $n = 18$, $p < 0.001$; Fig. 3C), indicating that modeled particle distributions can serve as a reasonable proxy for CH_4 transportation patterns, particularly in the metalimnion, where CH_4 may become trapped. The limits of this approach, with one main source in the deltaic area of the Rhône River, are noticeable close to S18 (LéXPLORE research platform) and suggest that (Fig. 3C), for that location, an additional sublittoral source is likely.

Isotopy and particle age to trace methane transformation

Our $\delta^{13}\text{C}$ - CH_4 measurements provided insights into methane sources and transformation (Fig. 4). In the epilimnion, average values ranged from -55.3‰ to -49.9‰ , with discrete depth measurement values extending from -56.5‰ to -49.5‰ (Fig. 4A and Supporting Information Fig. S7). As expected, the most depleted values were observed in stations near the Rhône River delta and along the nearshore (e.g., S2, S3, S7, and S10). An increase of $>4\text{‰}$ toward offshore

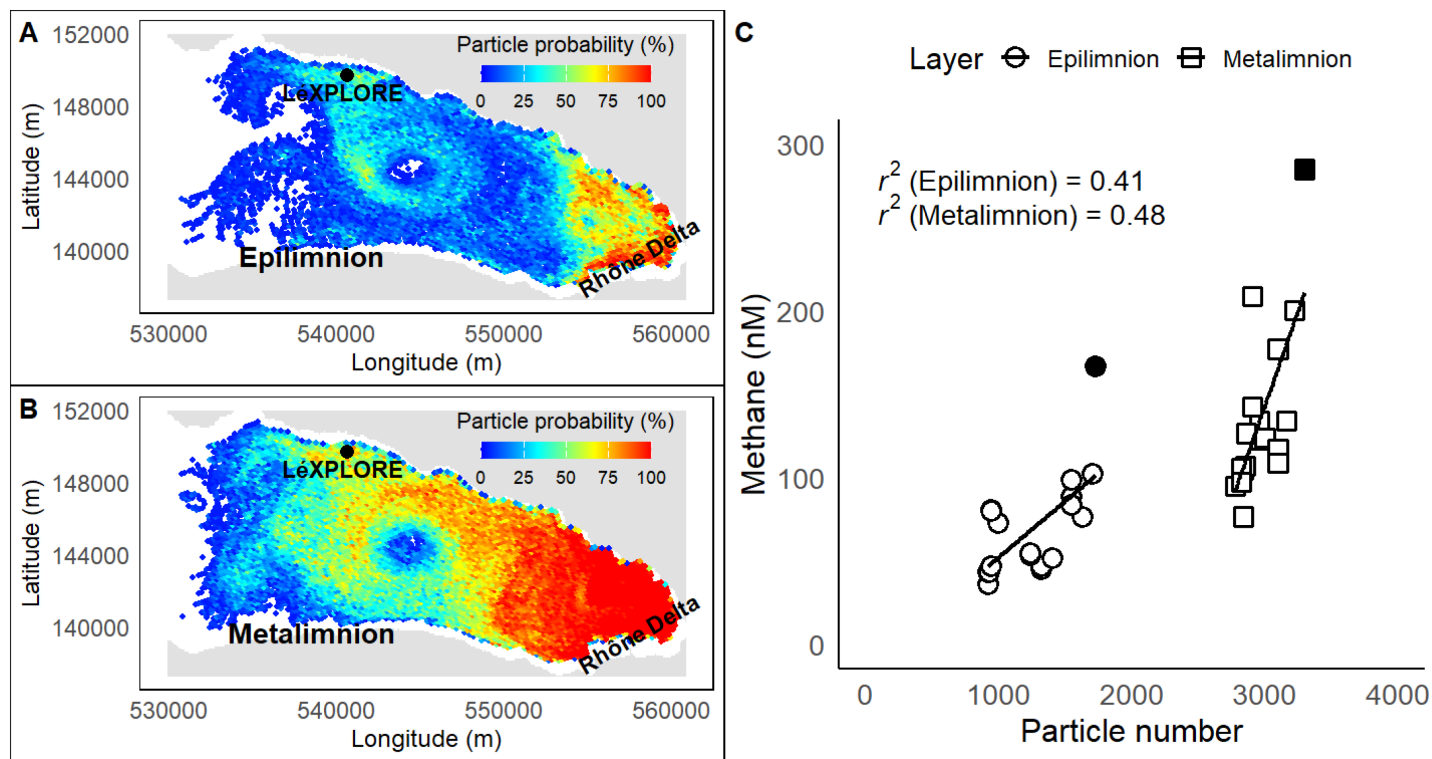


Fig. 3. Dominant deltaic water pathways obtained from the particle tracking model linked to dissolved CH_4 distribution across Lake Geneva. (A) and (B) show the probability maps of particles (%) reaching each location in the epilimnion (0–6 m) and metalimnion (6.1–20 m), respectively, between 28 June 11:30 and 29 June 17:30 (To visualize dominant transport pathways, particles released between 24 June 5:30 to 26 June 5:30 were tracked, see Supporting Information), with warmer colors (yellow to red) indicating a higher number of particles. The color scale is normalized for both layers for direct comparison of the patterns. (C) Correlation between observed depth-averaged methane (nM) and the simulated total number of particles that visited each of the 18 stations until their corresponding measurement times. Model performance metrics are: Epilimnion RMSE = 23.75 nM, SE = 5.76 nM, and Metalimnion RMSE = 35.83 nM, SE = 8.69 nM. These values are low relative to the maximum observed methane (~ 300 nM), reflecting a good model fit (see Supporting Information). The circles and squares represent epilimnion and metalimnion samples, respectively. The filled shape indicates the highest value at S18, near the LéXPLORE station.

stations, suggesting progressive isotopic enrichment. In the metalimnion, $\delta^{13}\text{C-CH}_4$ values were more variable (-49.1‰ to -39.1‰), with raw values spanning -53.7‰ to -30.5‰ (Fig. 4B and Supporting Information Fig. S7). Notably, the highest $\delta^{13}\text{C-CH}_4$ values (-47.1‰ to -30.5‰) were observed at 20 m depth, even at stations adjacent to the river delta.

Given the substantial $\delta^{13}\text{C-CH}_4$ enrichment observed in the metalimnion (Fig. 4B), we estimate CH_4 oxidation time (which can be considered as reaction time) using the Rayleigh fractionation model in combination with first-order reaction kinetics (see Supporting Information). Isotope-derived oxidation times range from 1 to 2.3 d in the epilimnion and 2.5–4.5 d in the metalimnion (Fig. 4C). These timescales are consistent with volumetric CH_4 oxidation rates of 22–26 $\text{nmol L}^{-1} \text{d}^{-1}$ reported in stratified lakes (Günthel et al. 2019) and with rates of 0–37 $\text{nmol L}^{-1} \text{d}^{-1}$ observed at the interflow in Lake Geneva (Khatun et al. 2024). When normalized to typical dissolved CH_4 concentrations ($\sim 100 \text{ nM}$), these rates correspond to effective first-order oxidation rate constants of $\sim 0.2\text{--}0.3 \text{ d}^{-1}$ (see Supporting Information), yielding characteristic oxidation times of $\sim 3\text{--}5 \text{ d}$. Shorter

epilimnetic oxidation times (lighter CH_4) may indicate rapid atmospheric evasion, whereas longer metalimnetic oxidation times (heavier CH_4) reflect extended residence of trapped CH_4 with limited vertical fluxes under higher DO concentrations (Supporting Information Fig. S3B).

Here, model-based particle age (a proxy of transport time) varies from 1 to 2.4 d near the delta to 3.97 d toward the lake center (Fig. 4D), similar across layers. No significant correlation is found between the averaged particle age and $\delta^{13}\text{C-CH}_4$ values. This weak correlation is tentatively explained by the simplified initial conditions of the particle release, both in space and time. However, looking at individual snapshots such as the one at 3.8 d (93 h, Supporting Information Fig. S8) shows a positive correlation in the metalimnion ($r^2 = 0.21$, $p = 0.0114$), suggesting aged deltaic CH_4 due to advection and recirculation despite the limitations indicated above. To evaluate such temporal discrepancies, we analyze hourly slopes for both layers when $\delta^{13}\text{C-CH}_4$ isotopes are station-specific and do not evolve along particle trajectories (Supporting Information Fig. S9). Only a small fraction of simulations showed statistically significant temporal trends

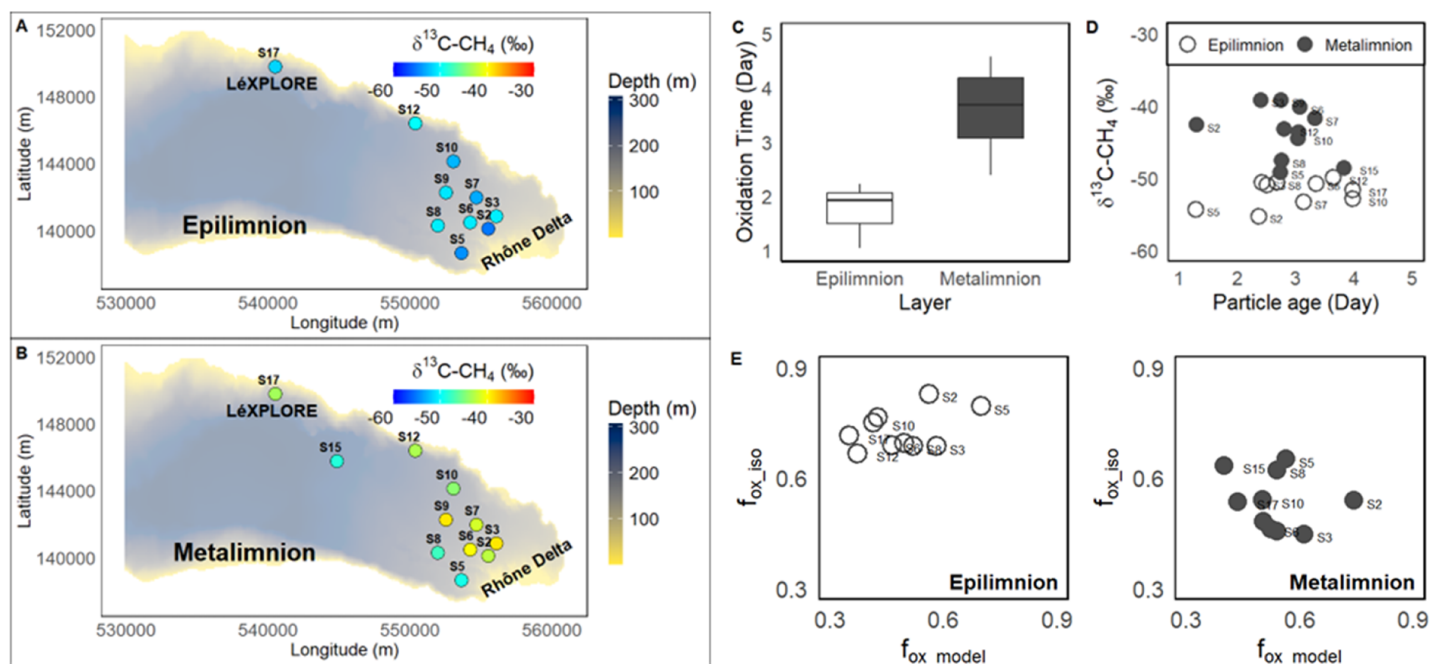


Fig. 4. Coupling stable carbon isotopes and particle age to trace methane transformation into stratified waters. Left panels: average $\delta^{13}\text{C-CH}_4$ value at each sampling station in the epilimnion (A) and metalimnion (B), respectively, illustrating spatial patterns in methane isotopic composition within each layer. The blue-red color scale indicates $\delta^{13}\text{C-CH}_4$ values in both layers, and the yellow-bluish scale indicates the bathymetry (m). (C) Boxplot showing isotope-derived oxidation time, $t_{\text{ox_iso}}$ (Day) in the epilimnion and metalimnion. The boxes represent the 25th to 75th percentiles, with the median indicated by the horizontal line. Whiskers extend to the smallest and largest values within 1.5 times the interquartile range. (D) Relationship between model-derived particle age, $t_{\text{ox_model}}$ (Day) derived from the particle tracking model and average $\delta^{13}\text{C-CH}_4$ values across stations. The particle age indicates the average travel time of particles to reach each station after their first release in the metalimnion near station S2. White and black circles represent epilimnion and metalimnion samples, respectively; station numbers are shown next to each point. (E) Relationship between isotope-derived methane fraction remaining ($f_{\text{ox_iso}}$) and transport-derived fraction remaining ($f_{\text{ox_model}}$) in the epilimnion and metalimnion. $f_{\text{ox_iso}}$ was estimated from $\delta^{13}\text{C-CH}_4$ using a Rayleigh fractionation model ($\delta^{13}\text{C}_0 = -60\text{‰}$, $\epsilon = -25\text{‰}$). $f_{\text{ox_model}}$ was derived from particle age assuming first-order decay with $k = 0.25 \text{ d}^{-1}$ (see Supporting Information). The comparison assesses consistency between isotope-inferred oxidation and transport-derived methane decay.

(4.8%, $p < 0.05$; Supporting Information Table S1) across both layers, indicating that $\delta^{13}\text{C}\text{-CH}_4$ variability is not governed by particle age alone. Additional processes not explicitly represented in the particle tracking framework, such as diffusion and microbial oxidation processes (Whiticar 1999) or other sources, may influence the observed isotopic patterns.

To further assess consistency between transport-derived and biogeochemical transformation indicators, we compare the first-order oxidation proxy from model-based particle age ($f_{\text{ox_model}}$) with the isotope-based indicator ($f_{\text{ox_iso}}$) in both layers (Fig. 4E). A weak positive relationship is observed in the epilimnion, whereas no clear correlation is found in the metalimnion.

Discussion

Stratification-driven hydrodynamics and lateral CH_4 transport are increasingly recognized as key drivers of methane accumulation in the oxygenated lake layers (Peeters and Hofmann 2021; Khatun et al. 2024; Liu et al. 2024). In Lake Geneva, earlier work ruled out OMP as the dominant source of metalimnetic CH_4 and favored laterally transported, sublittoral methane from the Rhône delta (Khatun et al. 2024). Despite excluding oxidation, pelagic production, and other littoral-pelagic transport, our model resolves key horizontal pathways of metalimnetic CH_4 using the high-resolution Sub-Ocean, which remain inaccessible to 1D models constrained to spatially averaged lateral diffusion alone. Because advection transport is orders of magnitude faster than lateral diffusion over kilometer scales (see Supporting Information), lateral CH_4 transport in Lake Geneva is dominated by circulation-driven advection, highlighting the need for a 3D hydrodynamic model. Higher CH_4 concentrations near the Rhône delta, combined with hydrodynamics, suggested northward advective transport. However, our previous model, based on limited data, could not fully explain high CH_4 concentrations at off-shore sites like LÉXPLORE, pointing to a possible sublittoral source and shorter-range transport. Our current model nevertheless provides spatial validation of our hypothesis, revealing elevated CH_4 maxima near the Rhône delta and along the southern shore (Figs. 2 and 3). Near-shore enrichment and depleted $\delta^{13}\text{C}\text{-CH}_4$ values further confirmed a littoral or deltaic origin rather than pelagic production (Hofmann et al. 2010; Donis et al. 2017; Grilli et al. 2023; Liu et al. 2024). We herein focus on the dominant transport pathways from the Rhône delta, without claiming to represent all CH_4 sources, while still providing mechanistic insight.

Our 3D hydrodynamic model supports that advective transport from the Rhône delta drives lateral flows, which are key in shaping long-range distributions of metalimnetic CH_4 across Lake Geneva, as previously suggested (Khatun et al. 2024). Particle tracking simulations show consistent east-west advection patterns and CH_4 accumulation aligned

with such deltaic influence (Figs. 2 and 3). These transport pathways match well with the spatial CH_4 measurements and modeled particle presence ($r^2 = 0.52$; Supporting Information Fig. S10), reinforcing the role of physical advection in shaping lake-wide CH_4 dynamics (Encinas Fernández et al. 2016; Grilli et al. 2023; Khatun et al. 2024). Thus, while not representing all CH_4 sources, our model shows that advective transport from a major deltaic input reproduces key features of the lake-wide CH_4 distribution and reveals physical controls unresolved by 1D models.

Interpreting isotopic data alongside the 3D hydrodynamic model remains challenging, and no direct correlation between $\delta^{13}\text{C}\text{-CH}_4$ and particle age is found. The temporal consistency of the slopes suggests that the processes controlling CH_4 isotopic composition relative to particle age are stable during the modeled period (Supporting Information Fig. S9). This stability implies limited bias in particle selection or changes in methane cycling dynamics. Vertical particle excursions, likely driven by stratification or internal waves, can facilitate the upward and downward movement of particles (Supporting Information Fig. S3). The advective Damköhler number (Da , used only as a diagnostic metric, Antenucci et al. 2013, see Supporting Information Fig. S11) remained below or near unity across most stations in both layers, suggesting comparable transport and transformation timescales. While the relationship between isotope and model-derived fractionation (Fig. 4E) suggests more coherent transport-oxidation coupling in the epilimnion than in the metalimnion, where more complex processes may weaken this relationship.

Epilimnetic $\delta^{13}\text{C}\text{-CH}_4$ values (-56.5‰ to -49.5‰) indicate a predominantly anaerobic sedimentary origin with limited isotopic enrichment (Conrad et al. 2011; Schubert et al. 2010, 2011), consistent with short CH_4 residence times and continued supply via lateral transport (Liu et al. 2024). Oxidation may be limited by ultraviolet (UV) exposure, nutrient limitation, and low methanotrophic activity in surface waters (Murase and Sugimoto 2005; Bastviken et al. 2008; Oswald et al. 2015).

Minor $\delta^{13}\text{C}\text{-CH}_4$ enrichment from the epilimnion (-56.5‰) to the upper metalimnion (-53.7‰) contrasts with more pronounced enrichment near 20 m depth (-30.5‰ ; Fig. 4). This likely reflects increased methane oxidation, facilitated by prolonged residence of deltaic CH_4 (Sollberger et al. 2014; Khatun et al. 2024). Since the metalimnion often aligns with the oxycline, oxygen or alternative electron acceptors may support methanotrophy (Sturm et al. 2016; Thottathil et al. 2019). Particle accumulation zones (e.g., circulation gyres) with increased turbidity and density may act as micro-niches that enhance CH_4 oxidation by increasing methanotrophic activity (Oswald et al. 2015; Mayr et al. 2020). These findings help explain the “methane paradox” in stratified lakes, where CH_4 persists in oxic layers due to lateral sublittoral input and gradual oxidation, which

leads to isotopic enrichment in the metalimnion (Boehrer and Schultze 2008; Wetzel 2001).

By combining real-time CH_4 and $\delta^{13}\text{C}\text{-CH}_4$ measurements with 3D particle-tracking, we show how circulation patterns can redistribute CH_4 across epilimnion and metalimnion, while outpacing metalimnetic oxidation along transport pathways. Importantly, we deliberately limited model runtime to maintain robustness and avoid uncertainties from prolonged simulations beyond CH_4 transformation timescales. A rough estimate of surface CH_4 fluxes ($0.1\text{--}30\text{ mg m}^{-2}\text{d}^{-1}$, see Supporting Information) suggests substantial evasion from the upper epilimnion ($\sim 0.5\text{ m}$) with notable spatial variability, corresponding to the lower limits reported earlier (Bastviken et al. 2004; Wik et al. 2016). Although flux estimation is not our primary goal here, because ebullition fluxes are not represented in the model, these findings further emphasize the need for laterally targeted investigations of CH_4 exchange during long-range horizontal transport. Overall, our study provides one of the first spatial validations of CH_4 transport, highlighting the need to resolve horizontal fluxes to improve the accuracy of methane budgets and understanding of cycling in deep stratified lakes.

Author Contributions

Santona Khatun and Marie-Elodie Perga formulated the central research problem. Santona Khatun, Roberto Grilli, Jérôme Chappellaz, and Marie-Elodie Perga conducted the field survey. Santona Khatun performed all laboratory analyses. Roberto Grilli and Jérôme Chappellaz carried out the SubOcean measurements and analyses. Abolfazl Irani Rahaghi and Damien Bouffard executed the hydrodynamic modeling analysis. Tomy Doda and Carsten J. Schubert assisted with data analysis and visualization. Santona Khatun drafted the manuscript with contributions from all co-authors. All authors have read and approved the final manuscript.

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Conflicts of Interest

None declared.

Data Availability Statement

The data supporting the findings of this study are available via Zenodo at <https://doi.org/10.5281/zenodo.17794709>.

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Supporting Information

Additional Supporting Information may be found in the online version of this article.

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