Local processes with a global impact: unraveling the dynamics of gas evasion in a step-and-pool configuration

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Received: 19 April 2023 – Discussion started: 21 April 2023
Revised: 29 June 2023 – Accepted: 9 July 2023 – Published: 11 August 2023

Abstract. Headwater streams are important sources of greenhouse gases to the atmosphere. The magnitude of gas emissions originating from such streams, however, is modulated by the characteristic microtopography of the riverbed, which might promote the spatial heterogeneity of turbulence and air entrainment. In particular, recent studies have revealed that step-and-pool configurations, usually found in close sequences along mountain streams, are important hotspots of gas evasion. Yet, the mechanisms that drive gas transfer at the water–air interface in a step-and-pool configuration are not fully understood. Here, we numerically simulated the hydrodynamics of an artificial step-and-pool configuration to evaluate the contribution of turbulence and air entrainment to the total gas evasion induced by the falling jet. The simulation was validated using observed hydraulic features (stage, velocity) and was then utilized to determine the patterns of energy dissipation, turbulence-induced gas exchange and bubble-mediated transport. The results show that gas evasion is led by bubble entrainment and is mostly concentrated in a small and irregular region of a few square decimeters near the cascade, where the local gas transfer velocity \( k \) peaks at \( 500 \text{ m d}^{-1} \). The enhanced spatial heterogeneity of \( k \) in the pool does not allow one to define a priori the region of the domain where the outgassing takes place and makes the value of the spatial mean of \( k \) inevitably scale-dependent. Accordingly, we propose that the average mass transfer velocity should be used with caution to describe the outgassing in spatially heterogeneous flow fields, such as those encountered in step-and-pool rivers.

1 Introduction

Headwater streams play a pivotal role in the biogeochemical functioning of river networks and regulate key ecosystem services associated with fluvial environments (Appling et al., 2018; Bernhardt et al., 2018). Furthermore, mountain streams not only transport matter and chemicals from the uplands to downstream waterbodies but are also able to exchange gaseous compounds with the overlying atmosphere (Butman and Raymond, 2011; Battin et al., 2009). Despite the small flow rates and reduced water–air interfaces that characterize low-order streams, the efficiency of gas transfer in high-energy mountain settings makes these waterbodies a significant source of greenhouse gases (such as \( \text{CO}_2 \) and \( \text{N}_2\text{O} \)) to the atmosphere. Thus, enhancing our understanding of gas evasion from headwater streams would have important implications for a broad range of fields in biology and environmental sciences (Kroeze et al., 2005; Crawford et al., 2014; Schelker et al., 2016; Marzadri et al., 2017; Horgby et al., 2019; Hall and Ulseth, 2020).

Yet, quantifying the cycling and release of greenhouse gases in low-order streams remains challenging, owing to the intertwined effect of different biotic and abiotic agents. In particular, the physical mechanisms responsible for gas evasion in river networks are often modulated by the hydrodynamic features of the flow field, which are in turn strongly impacted by the (micro)topography of the riverbed (Duvert et al., 2018; Botter et al., 2021; Rocher-Ros et al., 2019; Looman et al., 2021). Differently from floodplain rivers – which exhibit smooth hydraulic conditions – the steep slopes and irregular beds typical of mountain streams yield complex and heterogeneous flow patterns. In this setting, the free surface of the water flow can frequently break, owing to obstacles, bends or abrupt variations in the channel bottom.
Recent studies evidenced that local steps are important hotspots of gas evasion, to the point that in all the settings where steps are distinct from the stream’s morphology (e.g., in high-energy rivers or in streams originated by glacial processes), they control the overall network-scale outgassing (Vautier et al., 2020; Botter et al., 2022). While it has been argued that gas transfer associated with riverbed drops is promoted by the concurrence of high turbulence and bubble entrainment (see, e.g., Ulseth et al., 2019), there is limited knowledge of the physical processes that drive outgassing dynamics in local steps (see, e.g., Cirpka et al., 1993). Moreover, to date, the spatial patterns of energy dissipation and gas exchange in the plunging jet and the receiving pool of individual steps have not been analyzed. Consequently, the characteristic length scales of turbulence-induced and bubble-mediated gas transport in a step-and-pool configuration are still unknown. The characterization of the local and spatially heterogeneous nature of gas evasion in step-and-pool formations has important practical and theoretical implications. In particular, the spatial heterogeneity of gas exchange along a step constrains our ability to conceptualize such geomorphic elements by identifying a priori the size of the surface through which most of the gas exchange occurs (i.e., the step gas footprint) and the corresponding “effective gas transfer velocity” therein.

The present work aims at filling these knowledge gaps by combining experimental data and numerical modeling. In particular, here we reproduce the hydrodynamics of a simple step-and-pool geometry within an artificial flume using numerical simulations, which are then used to model the underlying gas exchange mechanisms taking place across the step. The numerical analysis allows us to (i) quantify the spatial variability in the gas transfer velocity, discerning the contributions of the turbulence and the entrained in-water bubbles, and (ii) identify the spatial patterns of energy dissipation and gas evasion in a step-and-pool configuration. The major implications of our findings for future gas transfer studies are also discussed.

2 Methods

2.1 In-flume experiment

An experiment to study the hydrodynamics of a step-and-pool configuration was carried out under controlled hydrodynamic conditions inside a horizontal, 6 m long plexiglass-made artificial flume with a rectangular section of width and height equal to 0.3 and 0.5 m, respectively. The water circulates through the channel via a constant head tank that maintains a steady discharge of 2 L s\(^{-1}\), which is accurately measured by a magnetic flowmeter.

A step-and-pool formation was artificially created in the flume by inserting a broad-crested weir and a tapered positive step (see Fig. 1). The height of the two elements was 25 and 9 cm, respectively. Owing to the difference in the water level upstream and downstream of the weir, a local step with a water height drop \(\Delta H\) equal to \(\approx 15\) cm was obtained. The pool, bounded by the weir and the step, had a depth of \(z_0\approx 11\) cm. The pool was long enough (100 cm, about 10 times the water depth) so that the hydrodynamics near the jet were not impacted by the presence of the downstream positive step. Finally, an orifice on the wall of the weir ventilated the air below the cascade jet to maintain atmospheric pressure therein.

We measured the water level along the step-and-pool configuration using a graduated hydrometric rod (accuracy \(\pm 0.1\) mm) mounted on a moving carriage. Measurements were taken every 2 cm along the middle axis of the flume, allowing us to reconstruct the free-surface profile in the region of interest.

Local flow velocity was assessed in some representative sections along the middle axis of the pool by two acoustic Doppler velocimeters (ADVs). To this aim, we used a Vectrino Profiler ADV (Nortek, Rud, Norway) to record the velocity near the bottom. The profiler provides observations of the three-component velocity, \(u_i\), with a resolution as fine as 1 mm over a 30 mm range placed 40 mm below the transmitter and an output rate of 100 Hz. For each section, we carried out a series of measures as follows: starting from the bottom, we repeatedly raised the ADV in 10 mm increments, recording velocities for 30 mm of water column each time. Since the ADV works correctly only when the transmitter is dipped in the water, the fluid volume close to the air–water interface (with a distance from the bed of more than 70 mm) was not analyzed. Therein, the velocity was assessed by a 2D SonTek ADV (Xylem Inc., Rye Brook, NY, USA). This ADV provides two-component velocity measures for a single point that corresponds to a control volume of 1 cm\(^2\), with a maximum sampling rate of 50 Hz. Measures of the horizontal components of the velocity were taken every 10 mm for 70 mm starting from the free surface.

To provide statistically consistent samples, the ADVs recorded the velocity signal for more than 30 s. During the tests, talcum powder was added to the water to achieve a signal-to-noise ratio (SNR) higher than 30 dB, thus minimizing the background noise recorded by the instruments.

2.2 Numerical simulation of the step-and-pool configuration

The in-flume experiment was numerically simulated using the software FLOW-3D Hydro 2022R1 (Flow Science Inc., Santa Fe, NM, USA), which adopts grids based on structured orthogonal cells to shape complex geometries using the fractional area–volume method (FAVOR). The evolution of the free surface is instead modeled using the volume-of-fluid (VOF) method (Hirt and Nichols, 1981), which can also track complex interfaces.
To simulate the hydrodynamic field of the step-and-pool 
configuration, the software solves the Reynolds-averaged 
Navier–Stokes equations along the Cartesian axes \( x_i \) coupled 
with the RNG (re-normalization group) \( e_k-\varepsilon \) scheme (\( e_k \) and 
\( \varepsilon \) being the turbulent kinetic energy per unit of mass and 
the turbulent kinetic energy dissipation rate, respectively), 
which resolves the turbulence at the sub-grid scale (Yakhot 
and Orszag, 1986).

The simulated dynamics in the pool included a full 
description of the bubble entrainment associated with the water 
flow. The air model used in this paper describes air transport 
in the water column according to the standard advection–
diffusion equation of a scalar with known concentration \( C \).
In the simulation, the air bubbles were assumed to be spheres 
with a radius, \( r \), equal to 0.5 mm, a value congruent with the 
analysis of cam-derived images taken during the experiment 
and in line with previous experimental studies (Woolf et al., 
2007; Klaus et al., 2022; Karn et al., 2016). The model al-
lowed us to quantify the entrained and escaping bubble fluxes 
across the free surface, \( q_B \).

In the numerical experiment, the cell size ranged from 
2 mm in the region where the jet plunged to 8 mm upstream 
of the broad crest weir. In total, the flume was discretized 
using approximately 7.5 million cells. The boundary condi-
tions were the following: (i) a steady inflow of 2.0 L s\(^{-1}\) up-
stream of the weir and (ii) free outflow downstream of the 
step. The no-slip velocity constraint was set on both the bot-
tom and channel sides, where the equivalent roughness was 
set at \( \delta_e = 5 \times 10^{-2} \) mm. Initially, fluid was at rest, and tran-
sient conditions were observed for less than 20 s before sta-
tionary conditions were reached in the domain.

2.3 Modeling gas exchange in step-and-pool 
configurations

The gas exchange between water and the atmosphere takes 
place through the thin separation layer at the interface be-
tween the two fluids. The exchanged volumetric gas flux, \( F \), 
is proportional to the difference in gas concentration between 
air and water and is described by Henry’s law as

\[
F = k \left( C_w - \frac{C_a}{K_H} \right), \quad (1)
\]

where \( k \) is the gas transfer velocity (representing the water 
depth equilibrated with the atmosphere in the unit of time);
\( C_w \) and \( C_a \) are the gas concentration in water and atmos-
phere, respectively; and \( K_H \) is the Henry constant deter-
mined for the specific exchange process under investigation. 
When both fluids are at rest, the exchange rate depends on the 
molecular diffusivity of the dissolved gas, \( D \), which slowly 
drives the absorption to (or the release from) the water col-
umn. Instead, in turbulent flowing water \( k \) is much higher 
than that induced by the molecular diffusivity. In this case, 
the eddies originating from turbulence continuously renew 
the water close to the free surface, enhancing the transfer of 
gas at the interface (Lamont and Scott, 1970; Katul and Liu, 
2017). Hereafter, the gas transfer velocity driven by turbulent 
mechanisms of this type is defined as \( k_T \).

However, in many real-world settings, air entrainment and 
bubbles can intensify gas exchange in heterogeneous flow 
fields, primarily by increasing the total exchange area be-
tween water and air. The bubble-mediated gas exchange is 
cepsulated here by the bubble-mediated gas transfer ve-
locity, \( k_B \). Therefore, in turbulent flowing waters where air 
entertainment takes place, the total gas transfer velocity \( k \) 
can be computed as the sum of velocities due to free-surface 
and bubble-mediated exchange (Klaus et al., 2022):

\[
k = k_T + k_B. \quad (2)
\]

The separation of the contributions of \( k_T \) and \( k_B \) to the total 
gas exchange in a step-and-pool configuration is one of the 
main contributions of this paper. The following subsections 
describe how these terms are computed in our numerical sim-
ulation.

2.3.1 Turbulence-induced air–water gas exchange in 
the flume

Existing estimates of gas exchange in the presence of turbu-
 lent flows are based on the idea of linking \( k_T \) with some key 
characteristic hydrodynamic quantity (e.g., the energy dis-
sipation). In mountain streams, with low water depths and 
high velocities, the theoretical schemes that are better suited 
to quantify \( k_T \) are those based on small-eddy models (Moog 
and Jirka, 1999b). In this framework, the renewal of the free 
surface of the water column is led by the small-scale vor-
tices originated by the turbulence near the interface, which 
in turn are controlled by the rate of dissipation per mass unit 
of the turbulent kinetic energy, \( \varepsilon \). According to this model,
$k_T$ scales with $\varepsilon$ as follows:

$$k_T = \alpha_T \varepsilon^{0.5} (v \varepsilon)^{0.25}, \quad (3)$$

where $\alpha_T = 0.2 - 0.4$ is a calibration factor, and $v \approx 1 \times 10^{-6} \text{m}^2\text{s}^{-1}$ is the kinematic viscosity of water. Moreover, in Eq. (3), $Sc = v/D$ is the Schmidt number, which expresses the link between kinematic and molecular diffusivity. The nature of the processes described by Eqs. (1) and (3) is inherently local, and they occur at the Batchelor scale (Batchelor, 1959), which is defined as $\lambda_B = \eta/\sqrt{Sc}$, where $\eta = (v^3/\varepsilon)^{1/4}$ is the Kolmogorov length, i.e., the characteristic length of the microvortexes. In running waters, $Sc > 1$, the spatial scale of the gas transport is thus smaller than that of the smallest eddies generated by turbulence in its energy cascade, that is, the process of the energy transfer from the large-scale eddies to microvortexes (Tennekes and Lumley, 1972).

Previous experimental studies have demonstrated that the exponent $-0.5$ in Eq. (3) applies to cases in which the free surface is smooth, while the same exponent may decrease to $-0.67$ if the free surface is riffled (Zappa et al., 2007; Jähne et al., 1987). It should be noted that, in the general case, the gas exchange also depends on the fluid temperature, which affects both $v$ and $D$. However, since the present work aims at reproducing a short portion of a stream in which the water temperature is nearly constant, we neglect the thermal effect on $D$ and $v$ so that $Sc$ is assumed to be constant, and the gas transfer velocity $k_T$ scales with $\varepsilon^{0.25}$.

Nevertheless, in-field estimations of gas transfer velocity are usually performed at the reach scale based on spatially averaged quantities under the assumption of uniform flow. Under the above circumstances, the mean turbulent kinetic energy dissipation rate $\overline{\varepsilon}$ (hereinafter the overline applies to spatially averaged quantities) can be expressed as (Ulseth et al., 2019; Hall and Ulseth, 2020; Hall and Madinger, 2018; Raymond et al., 2012)

$$\overline{\varepsilon} = g \overline{U} \overline{s}, \quad (4)$$

with $g$ the gravity acceleration, $\overline{U}$ the cross-sectional flow velocity and $\overline{s}$ the average bed slope. It should be noted that the proposed scheme is suitable if the turbulence generated at the bottom is the main driver of the overall intensity of the turbulence in the water flow. In headwater streams, instead, riverbed heterogeneity, partially emergent boulders and small cascades can be important additional turbulence sources (Moog and Jirka, 1999a; Botter et al., 2022). In the particular setup analyzed in this study, the plunging jet of a cascade dissipates its energy by inducing high $\varepsilon$ in the outermost fluid layer, thereby preventing the use of Eq. (4).

Consequently, $k_T$ is calculated based on the computed value of $\varepsilon$ near the free surface, using Eq. (3) with $\alpha_T = 0.4$ and standardizing the result with a Schmidt number equal to 600.

### 2.3.2 Bubble-mediated gas exchange

In this work, while dealing with bubble-mediated gas exchange we focus on the so-called “kinematic bubble effect” (Liang et al., 2013). According to our approach, the amount of gas transferred from the water to the bubbles (or vice versa) is estimated using the following three-step procedure: first we determine the reference bubble lifetime, $T_B$ (i.e., the time spent by the bubble within the water column when it reaches the surface; Step A); then, we calculate the gas transferred across the bubble interface according to the difference in concentration compared to the surrounding bulk flow along the bubble flow path (Step B); finally, the total gas transfer velocity is estimated by considering the total amount of bubbles involved in the process (Step C).

- **Step A.** A reasonable estimation of the bubble lifetime, $T_B$, is given by

$$T_B = \alpha_B \frac{z_0}{u_B}, \quad (5)$$

where $\alpha_B$ is an $O(1)$ coefficient, and $u_B$ is the bubble rise velocity. Here, $u_B$ was estimated using the model proposed by Woolf (1993):

$$u_B = \begin{cases} 0.172 r^{1.28} g^{0.76} v^{-0.56} & \text{if } r \leq 0.82 \text{ mm} \\ 0.25 \text{ m s}^{-1} & \text{if } r > 0.82 \text{ mm} \end{cases}. \quad (6)$$

- **Step B.** During the time $T_B$, any bubble is able to exchange gas with the surrounding water as a function of $u_B$ and the Reynolds number $Re_B = 2 u_B r / v$. In particular, the velocity of gas exchange, $j$, through a bubble is given by the following expression (valid for $Re_B > 10$):

$$j = \sqrt{\left(1 - \frac{2.89}{\sqrt{Re_B}}\right) \frac{2 D u_B}{\pi r}}. \quad (7)$$

The velocity of gas exchange is functional to the computation of the gas equilibration time constant of the bubble, $T_g$, which defines the timescale of the gas transfer process and is given by

$$T_g = \frac{r}{3 j \beta}. \quad (8)$$

where $\beta$ is the Ostwald solubility coefficient, which is the ratio between the volume of absorbed gas and the volume of absorbing liquid for a given condition of pressure and temperature.

- **Step C.** The gas transfer velocity induced by bubbles, $k_B$, can be calculated assuming a first-order reaction model which accounts for the time-dependent changes in gas concentration within the drifting bubble:

$$k_B = \frac{q_B \beta}{\beta} \left(1 - e^{-\frac{r_B}{T_B}}\right), \quad (9)$$

with $q_B$ the bubble flux per unit area.
In this application, $k_B$ was calculated by combining the above equations, as detailed in what follows. The bubble rise velocity was calculated assuming a radius $r$ of 0.5 mm using Eq. (6) (leading to $u_B = 0.133 \text{ m s}^{-1}$). Then, the bubble lifetime $T_B$ was calculated setting $\alpha_B$ to 1.0 in Eq. (5) ($T_B \approx 0.83 \text{ s}$), and the Reynolds number was computed based on its definition ($Re_B = 133$). After that, assuming that in the case of $CO_2 D = 1.6 \times 10^{-9} \text{ m}^2 \text{s}^{-1}$ and $\beta = 0.94$, the bubble’s transfer velocity, $j$, and the equilibration time constant, $T_g$, were estimated via Eqs. (7) and (8) ($j \approx 0.5 \text{ mm s}^{-1}$ and $T_g = 0.34 \text{ s}$). Finally, $k_B$ was obtained from Eq. (9) using the local flux $q_B$ computed by the numerical code. Given the uncertainty in the value of $T_B$ in this particular experiment, a range of values for the gas transfer velocity induced by bubbles was derived by varying $\alpha_B$ in the interval (0.5, 2.0).

3 Results

3.1 Performance of the hydrodynamic model

To assess the reliability of the hydrodynamic model, the depths and velocities measured in the pool were compared to the numerical solution provided by the model. Figure 2a shows the comparison between the observed and modeled water levels along the middle axis of the flume. The water stage ranged from 10.5 in the pool to 12.0 cm downstream of the plunging jet, whereas the backwater upstream of the plunge had higher water depths. The modeled free surface fit the experimental levels in the pool; over the broad-crested weir; and, with lower accuracy, on the cascade. Therein, however, the measures taken with the rod were sub-optimal owing to observed abrupt variations in the water level. Further, Fig. 2a shows the model’s ability to reproduce the module of the velocity vector, $|U|$. The results highlight a non-uniform flow distribution along the vertical direction within the pool and downstream of the cascade.

Figure 2b reports the profile of the time-averaged longitudinal velocity ($u_x$) in three sections located at 10 (A), 20 (B) and 30 cm (C) downstream of the plunging jet. The velocities estimated by the ADV campaign were rather scattered, likely because of the strong fluctuations induced by the turbulent vortexes, and systematically underestimated close to the bottom. Nevertheless, the observed trends were consistent with the numerical solution of the hydrodynamic model. In all cases, the maximum values of ($u_x$) were close to the bottom and swiftly decreased downstream of the cascade (from approximately 0.75 m s$^{-1}$ (A) to 0.45 m s$^{-1}$ (C)). Moreover, the ($u_x$) decreased in the vertical direction, with a null velocity at 0.45 $z_0$ for Sect. A and 0.50–0.55 $z_0$ for Sects. B and C. Negative values are observed at a higher vertical distance from the channel bed, $z$. Consequently, the maximum backflow was observed near the free surface.

The three examples of velocity profiles pointed out the persistence of a large, counterclockwise eddy structure. The jet plunging in the pool also formed a clockwise vortex in the backwater region, as clearly indicated by the streamlines reported in Fig. 2c. During the experiment, air bubbles entrained by the water jet were visible within a 30 cm wide portion of the counterclockwise eddy and in the backwater region (blue circles in Fig. 2d). The sizes of most of the bubbles highlighted in the picture were in the range $r = 0.5–1.0 \text{ mm}$, while it was hardly feasible to detect smaller bubbles.

3.2 Gas evasion produced by the step-and-pool configuration

The simulated spatial patterns of total turbulent kinetic energy dissipation rate were highly heterogeneous (Fig. 3a). As expected, the water spilled from the weir dissipated almost all its energy within the region of the two large eddies. However, the highest values of $\varepsilon$ were found in the area where the water jet plunges. Therein, $\varepsilon$ was higher than $2.0 \text{ m}^2 \text{s}^{-3}$ at the free surface, with much smaller values – of the order of $0.1 \text{ m}^2 \text{s}^{-3}$ – near the bottom of the flume. The decrease in $\varepsilon$ with depth was superlinear (see inset of Fig. 3a). Therefore, overall, the first centimeters of the water column dissipated a significant amount of the jet energy. Moreover, the simulated patterns of $\varepsilon$ indicated that, in a small superficial region of the fluid mass with a size of $120 \text{ cm}^2$ (which is comparable with the footprint of the cascade in the pool), the energy dissipation rate was 1 order and at least 2 orders of magnitude larger than that estimated in the backwater region and downstream of the cascade, respectively.

The recirculating zones were also able to exchange gas with the overlying atmosphere as they trapped the air dragged by the falling jet (see Fig. 3b). The two large vortexes confined the air in some low-velocity regions located in the middle of the water column and swept the bubbles near the bottom and the free surface. According to the numerical model, the maximum bubble concentration was found close to the plunging jet ($C \approx 50 \text{ mg L}^{-1}$), and only a small number of bubbles were predicted at the edges of the region of the observed bubbles. Despite the steady-flow condition achieved (the global air mass in the pool remained almost constant during the simulation), the air entrainment spatial distribution varied significantly over time (see the Video supplement). The magnitude of the bubble flux, $q_B$, mirrored the patterns of air concentration (Fig. 3c); $q_B$ was highly heterogeneous in the pool, and most air bubbles escaped by bursts in big spots within the first 10 cm downstream of the cascade and the first 5 cm of the backwater region. In contrast with $\varepsilon$, the maximum value ($q_B \approx 5.0 \text{ mm s}^{-1}$) was not found at the cascade, where the air was entrained, but a few centimeters downstream of it.

The gas evasion induced by the step resulted from the superposition of the effect of turbulence and entrained bubbles. Figure 4a shows the spatial distribution of $k_T$. We also averaged $k_T$ along $y$ direction ($k_T$) and reported its longitudinal pattern along the step-and-pool configuration. Owing to the close connection between the gas exchange velocity and $\varepsilon$,
$\kappa_T$ reaches a maximum of $50 \text{ m d}^{-1}$ near the plunging jet. In this region of enhanced dissipation, local $\kappa_T$ values peaked at $100 \text{ m d}^{-1}$; such a value was almost 1 order of magnitude larger than those observed downstream of the pool, where $\kappa_T$ progressively decreased moving away from the jet. Conversely, in the backwater region $\kappa_T$ was nearly uniform, and $\kappa_T$ did not exceed $20 \text{ m d}^{-1}$.

In the region of the domain in which the bubble flow was enhanced, we estimated $\kappa_B$ values which were almost an order of magnitude higher than those computed for the turbulence-driven gas transfer velocity, $\kappa_T$ (Fig. 4b). In particular, $\kappa_B$ locally peaked at $500 \text{ m d}^{-1}$, while the highest values of $\kappa_B$ exceeded $100 \text{ m d}^{-1}$ — about twice the maximum value of $\kappa_T$.

The patterns of $\kappa_T$ and $\kappa_B$ were not strongly correlated in space and both showed pronounced peaks. Consequently, the relative contribution of bubbles and turbulence to the total gas evasion showed a heterogeneous spatial trend. Figure 4c reports the pattern of the dominance ratio $\chi = \kappa_B / \kappa_T$ along the pool where bubble entrainment was observed. The plot indicates that bubbles dominated the exchange rate ($\chi > 1$) in the central portion of the bubble region. In contrast, turbulence led to the outgassing ($\chi < 1$) near the cascade (where $\varepsilon$ was maximum) and at the edges of the pool (where air concentration was low).
4 Discussion

The important contribution of headwater streams to the global emissions of climatically relevant gases is modulated by the topography of the riverbed, which determines the steep, tortuous and irregular nature of water flow paths in high-energy settings (Duvert et al., 2018; Wallin et al., 2011; Marx et al., 2017). For example, small-scale heterogeneity of channel forms promotes rippled air–water interfaces that enhance the evasion of greenhouse gases. The sequences of step-and-pool configurations typical of mountain streams are plastic examples of small-scale geomorphic elements of rivers that control gas exchange processes at the water–air interface, eventually bearing a relevant impact on the chemical equilibrium of the atmosphere (Botter et al., 2022). Sharp discontinuities of the riverbed of the types observed in steps and small cascades cause a sudden transfer of potential energy to kinetic energy of the water flow. The water jet impacting the pool or the downstream portion of the riverbed then loses a large amount of its energy, roughly corresponding to the height difference between the water levels upstream and downstream of the cascade (i.e., $\Delta E \approx g \Delta H$, where $\Delta E$ is the dissipated energy per unit of fluid mass; see Botter et al., 2022). The dissipation of energy occurs within a few cen-
of the air that overlies the pool. The air bubbles in the flume falling jet is responsible for the entrapment and entrainment does not exceed a few centimeters near the cascade.

The overall result is that the turbulence depletes indicatively of values (colored area) is determined by assuming $\alpha$ to be equal to 0.2 and 0.6. (b) Field of $k_B$ and transverse-averaged exchange velocity $\bar{K}_B$. The solid line is estimated by Eq. (9) with $\alpha_B = 1$, while the range of values (colored area) is determined by assuming $\alpha_B$ to be equal to 0.5 and 2.0 (c) The $\chi = k_B/\bar{K}_T$ ratio within the pool.

In this area, the overall transfer velocity $k = k_T + k_B$ reaches a local maximum of about 500 m$^{-1}$, with transverse-averaged values of the mass transfer rate equal to $\bar{K} = 120$ m$^{-1}$. While we recognize that such values of $k$ are quite high – especially in light of the uniformity of the geometry in our flume – we expect that the outgassing induced by steps in mountain streams could be even larger owing to the higher values of $\Delta H$ (Natchimuthu et al., 2017; Schneider et al., 2020; Botter et al., 2022) and the enhanced heterogeneity of the bed geometry, which arguably intensify turbulence and air entrainment (Vallé and Pasternack, 2006; Vautier et al., 2020).

Interestingly, in the bubble region of the pool, the potential outgassing, i.e., the surface integral of Eq. (1) for a constant and unit air–water concentration difference, induced by bubble-mediated transport is 2.25 times higher than the corresponding outgassing implied by turbulence alone. Some caution is needed when transposing this numerical result to real-world settings, as the outgassing driven by bubbles may depend not only on the step geometry but also the size and distribution of the bubbles (see Eqs. 6–8). In our simulation, we used a fixed bubble radius, which was set to represent the size of the visible bubbles observed during the experiment. Thus, we have neglected the role of the microbubbles in light of the fact that the mass fraction associated with small bubbles is expected to be negligible (Garrett et al., 2000).

Although this analysis is based on several simplifying assumptions, we believe that the orders of magnitude of the underlying processes are properly captured by the model, as space-time heterogeneity in bubble size should not significantly impact the estimated flux of entrained air. Further, we suggest that this result could also apply to different settings, as white waters are observed not only in local steps but also in turbulent reaches where high Froude numbers, macroroughness and emerging stones can promote diffusive entrainment of air. Accordingly, we propose that most of the observed gas evasion in high-energy streams is likely bubble-mediated, in line with previous experimental and modeling studies (Ulseth et al., 2019; Vautier et al., 2020; Klaus et al., 2022). Therefore, we suggest that more effort should be directed to adequately modeling entrained air and gas transfer through bubbles in high-energy streams, where channel bottom morphology causes ripple-free surfaces.

Moreover, the key role of the bubbles in gas evasion from high-energy streams suggested by our results raises two issues about the correct procedure to define the value of $k$ for a given stream reach. First of all, our numerical simulation indicates that the standardization of $k$ should be done not only in terms of $Sc$ but also in terms of $\beta$ (and $D$), as suggested in previous studies (Woolf et al., 2007; Hall and Ulseth, 2020; Klaus et al., 2022; Hall and Madinger, 2018). Consequently, some caution is necessary when gas evasion

Figure 4. Gas exchange velocity in the pool. (a) Field of $k_T$ and transverse-averaged exchange velocity $\bar{K}_T$. The solid line is estimated by Eq. (3) with $\alpha_T = 0.4$, while the range of values (colored area) is determined by assuming $\alpha_T$ to be equal to 0.2 and 0.6. (b) Field of $k_B$ and transverse-averaged exchange velocity $\bar{K}_B$. The solid line is estimated by Eq. (9) with $\alpha_B = 1$, while the range of values (colored area) is determined by assuming $\alpha_B$ to be equal to 0.5 and 2.0 (c) The $\chi = k_B/\bar{K}_T$ ratio within the pool.

In addition, the velocity increase observed within the falling jet is responsible for the entrainment and entrainment of the air that overlies the pool. The air bubbles in the flume experiment extended for almost 30 cm downstream of the jet. However, the numerical simulation indicates that most of the air is exchanged only in the first 10 cm near the jet.

In this area, the overall transfer velocity $k = k_T + k_B$ reaches a local maximum of about 500 m$^{-1}$, with transverse-averaged values of the mass transfer rate equal to $\bar{K} = 120$ m$^{-1}$. While we recognize that such values of $k$ are quite high – especially in light of the uniformity of the geometry in our flume – we expect that the outgassing induced by steps in mountain streams could be even larger owing to the higher values of $\Delta H$ (Natchimuthu et al., 2017; Schneider et al., 2020; Botter et al., 2022) and the enhanced heterogeneity of the bed geometry, which arguably intensify turbulence and air entrainment (Vallé and Pasternack, 2006; Vautier et al., 2020).

Interestingly, in the bubble region of the pool, the potential outgassing, i.e., the surface integral of Eq. (1) for a constant and unit air–water concentration difference, induced by bubble-mediated transport is 2.25 times higher than the corresponding outgassing implied by turbulence alone. Some caution is needed when transposing this numerical result to real-world settings, as the outgassing driven by bubbles may depend not only on the step geometry but also the size and distribution of the bubbles (see Eqs. 6–8). In our simulation, we used a fixed bubble radius, which was set to represent the size of the visible bubbles observed during the experiment. Thus, we have neglected the role of the microbubbles in light of the fact that the mass fraction associated with small bubbles is expected to be negligible (Garrett et al., 2000).

Although this analysis is based on several simplifying assumptions, we believe that the orders of magnitude of the underlying processes are properly captured by the model, as space-time heterogeneity in bubble size should not significantly impact the estimated flux of entrained air. Further, we suggest that this result could also apply to different settings, as white waters are observed not only in local steps but also in turbulent reaches where high Froude numbers, macroroughness and emerging stones can promote diffusive entrainment of air. Accordingly, we propose that most of the observed gas evasion in high-energy streams is likely bubble-mediated, in line with previous experimental and modeling studies (Ulseth et al., 2019; Vautier et al., 2020; Klaus et al., 2022). Therefore, we suggest that more effort should be directed to adequately modeling entrained air and gas transfer through bubbles in high-energy streams, where channel bottom morphology causes ripple-free surfaces.

Moreover, the key role of the bubbles in gas evasion from high-energy streams suggested by our results raises two issues about the correct procedure to define the value of $k$ for a given stream reach. First of all, our numerical simulation indicates that the standardization of $k$ should be done not only in terms of $Sc$ but also in terms of $\beta$ (and $D$), as suggested in previous studies (Woolf et al., 2007; Hall and Ulseth, 2020; Klaus et al., 2022; Hall and Madinger, 2018). Consequently, some caution is necessary when gas evasion...
extrapolating observed reach-wise mass transfer rates could be appropriate only in a limited subset of reaches that share the same degree of heterogeneity of the reach where the experiments were performed. Based on the above arguments, we propose that the use of the mass transfer rate, $k$, should be used with caution when the heterogeneity of the flow field controls the fraction of mass evaded into the atmosphere, as in our step-and-pool configuration. In these cases, the mass transfer rate could be highly site-specific, and its extrapolation to different settings could be feasible only following a detailed analysis of the evasion produced by the constitutive geomorphic elements of the focus reach (including steps and cascades) (Botter et al., 2022).

5 Conclusions

In this study, we have numerically simulated the hydrodynamics of a step-and-pool configuration, which were also reproduced in a laboratory-designed flume experiment. Computational values of key hydrodynamic parameters that control the gas transfer between water and the atmosphere were then used to estimate the spatial patterns of gas transfer velocity and the contributions to the gas evasion due to turbulence and air entrainment. Our simulation shows that the cascade downstream of the step dissipates energy and entraps air in a small, irregular region near the chute (about 3 dm$^2$), in which the mass transfer velocity $k$ is locally very high (500 m$^{-1}$). This result indicates that gas evasion in step-and-pool configurations may be a very local process, taking place within a few square decimeters of the plunging jet. The numerical simulation also suggests that bubble-mediated gas exchange dominates the turbulence-induced outgassing, reinforcing the idea that gas evasion in mountain streams could be mainly driven by bubbles and white waters. The observed heterogeneity of $k$ – the pattern of which is linked to the specific geomorphic features of the step under investigation – raises concerns about the ability of traditional metrics such as the mass transfer rate to quantify gas evasion from step-and-pool configurations and contrast the outgassing potential of different morphologically heterogeneous streams.

Data availability. Data supporting the findings of this study are openly available in Peruzzo et al. (2023) at http://researchdata.cab.unipd.it/id/eprint/619.

Supplement. Animation of air entrainment is included in the Supplement. The supplement related to this article is available online at: https://doi.org/10.5194/bg-20-3261-2023-supplement.

Author contributions. Conceptualization: PP, ND and GB. Methodology: PP, MC and GB. Investigation: MC. Data curation: MC. Software: PP. Formal analysis: PP and GB. Visualization: PP. Writing –
original draft preparation: PP. Writing – review and editing: ND and GB. Funding: GB. Supervision: GB.

Competing interests. The contact author has declared that none of the authors has any competing interests.

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Financial support. This research has been supported by the H2020 European Research Council (grant no. 770999).

Review statement. This paper was edited by Gabriel Singer and reviewed by two anonymous referees.

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