Supplementary materials

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Table 1: Experimental conditions imposed at the source. *: linear stratification experiment, in which the tank is filled with salt water using the double bucket technique. Values at the LNB (C) are calculated using a model of turbulent jet (Carazzo et al., 2008) with conditions imposed at the source (N).

<table>
<thead>
<tr>
<th>Exp.</th>
<th>ΔT₀ (K)</th>
<th>ΔS₀ (psu)</th>
<th>C₀ (g/g)</th>
<th>ρ₀ (kg m⁻³)</th>
<th>Q₀ (m³ s⁻¹)</th>
<th>ΔT_u (K)</th>
<th>ΔS_u (psu)</th>
<th>ΔC_u (g/g)</th>
<th>R_p</th>
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<td>0</td>
<td>1.2</td>
<td>1 × 10⁻²</td>
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<td>2</td>
<td>0</td>
<td>10</td>
<td>0.025</td>
<td>1013.6</td>
<td>4.1 × 10⁻⁵</td>
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<td>993.2</td>
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<td>4.6</td>
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Table 2: Physical characteristics of event plumes. H_d: thickness of the cloud thickness; H_l: thickness of the underlying column of seawater; ΔT_max: maximum temperature anomalies observed; ΔC_max: maximum particle anomalies observed; T: residence time; n_obs: number of layers observed. References: (1): Baker et al. (1987); (2): Lupton et al. (1989); (3): Massoth et al. (1994); (4): Baker et al. (1989); (5): Nogiri et al. (1989); (6): Coale et al. (1991); (7): Gamo et al. (1993); (8): Baker et al. (1995); (9): Massoth et al. (1995); (10): Baker et al. (1998); (11): Feely et al. (1998); (12): Kelley et al. (1998); (13): Massoth et al. (1998); (14): Lupton et al. (1998); (15): Baker et al. (1999); (16): Edmonds et al. (2003); (17): Murton et al. (2006); (18): Baker et al. (2011); †possible event plume; ‡thickness estimated by grouping individual layers.

<table>
<thead>
<tr>
<th>Event</th>
<th>Location</th>
<th>Diameter (km)</th>
<th>H_d (m)</th>
<th>H_l (m)</th>
<th>ΔT_max (K)</th>
<th>ΔC_max (g L⁻¹)</th>
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<th>References</th>
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<td>700</td>
<td>300</td>
<td>0.28</td>
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<td>1</td>
<td>(1; 2; 3)</td>
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<tr>
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<td>16</td>
<td>600</td>
<td>400</td>
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<td>4.37 × 10⁻⁴</td>
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<td>(4; 3)</td>
</tr>
<tr>
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<td>0.03</td>
<td>N/A</td>
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<td>(5)</td>
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<td>800</td>
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</tr>
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<td>(18)</td>
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<td>N/A</td>
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<td>(18)</td>
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</table>
Supplementary Figure 1: (A) Comparison of plume (solid line) and ambient (dashed line) density profiles in our experiments (top) and in nature (bottom). Calculations are made with a model of turbulent jet (Carazzo et al., 2008). (B) Comparison of plume height as a function of time for two experiments made at $N^2 = 0.38 \text{ s}^{-2}$ with a linear stratification (red curve - exp. 6) and a single density interface (blue curve - exp. 4). The insets show two photographs of the experiments at $t = 100 \text{ s}$. (C) Theoretical predictions of plume velocity and radius for two experiments made at the same source conditions but with a linear stratification (red curve) and a single density interface (blue curve). These comparisons confirm that both lab-scale and natural plumes are qualitatively similar, and that the two-layer and linear density stratification give identical quantitative results.
A. Diffusivity for the particles

Classical experiments involving hot particle-laden fluids have been carried out using small particles ($3 - 20 \mu m$) with a low Brownian diffusion coefficient (Green, 1987; Huppert et al., 1991; Chen, 1997; Hoyal et al., 1999), which is given by

$$\kappa_p = \frac{k_0 T_a}{3\pi \rho_f \nu d_p^3},$$  \hspace{1cm} (A.1)

where $k_0$ is the Boltzmann constant, $T_a$ is the absolute temperature, $d_p$ is the particle diameter, $\rho_f$ is the interstitial fluid density, and $\nu$ is the kinematic viscosity. In our experiments, the solid fraction consists of non-Brownian particles ($d_p = 300 \mu m$) but the hydrodynamic diffusivity, which refers to the bulk random-walk motion of groups of particles in response to gradients in flow velocity and concentration, can be estimated from well-established theoretical and experimental studies of particle diffusion (e.g., Leighton and Acrivos, 1987a; Davis, 1996; Da Cunha and Hinch, 1996). For spherical particles this diffusivity is given by

$$\kappa_p = k \dot{\gamma} d_p^2 \epsilon_p^n,$$  \hspace{1cm} (A.2)

where $\epsilon_p$ is the particle concentration, $\dot{\gamma}$ is the local shear strain rate, and $k$ and $n$ are constant coefficients, which vary from one study to another ($k = 0.0075$ & $n = 1$ in Da Cunha and Hinch (1996), $k = 0.33$ & $n = 2$ in Davis and Leighton (1987); Acrivos et al. (1993), $k = 0.5$ & $n = 2$ in Leighton and Acrivos (1987a)). Hydrodynamic diffusion can arise under both low-Reynolds number (Leighton and Acrivos, 1987a,b; Schaufinger et al., 1990; Acrivos et al., 1993) and high-Reynolds number conditions (Solomatov et al., 1993; Solomatov and Stevenson, 1993; Rampall and Leighton, 1994; Huppert et al., 1995; Ziskind and Gutfinger, 2002). In our experiments, based on the rate of propagation of the water-rich layers we infer $\dot{\gamma} \sim 0.01 \text{ s}^{-1}$ (Carazzo and Jellinek, 2013), which leads to $\kappa_p = 10^{-14} - 10^{-13} \text{ m}^2 \text{ s}^{-1}$ for a range of $k$ and $n$ found in the literature.

A minimum bound for $\kappa_p$ in layered hydrothermal clouds is given by the Brownian diffusion coefficient. For $0.5 - 50 \mu m$ particles, Eq. (A.1) gives $\kappa_p = 8 \times 10^{-15} - 8 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$. From our experiments and well-established theoretical and experimental studies of particle diffusion, a more appropriate diffusivity is a hydrodynamic value (Davis, 1996; Carazzo and Jellinek, 2013). However, because there is no adequate observational constraints on the local particle concentration ($\epsilon_p$) and rate of shear across each PBL ($\dot{\gamma}$) in layered hydrothermal clouds, which enter both in a significant manner in Eq. (A.2), we cannot estimate the hydrodynamic diffusivity for the natural case. Comparing the Brownian and hydrodynamic diffusion coefficient in our experiments, we argue that a plausible hydrodynamic value in the natural case may be 2 to 4 orders of magnitude larger than the Brownian value, which still satisfies the instability condition given in Eq. (1) of the paper.

In spite of the uncertainties on $\kappa_p$, the criterion for the onset of particle diffusive convection is largely attained in both our experiments and hydrothermal clouds (Eq. 1 of the paper). Furthermore, the effective value of $\kappa_p$ has no influence on the quantitative predictions made with our sedimentation model (Eqs. 4 & 5 of the paper) since the rate of change in particle content from the cloud is controlled by the diffusivity of the fastest diffusive substance (i.e., temperature), whose value is well known ($\kappa_T = 10^{-7} \text{ m}^2 \text{ s}^{-1}$ in our experiments, and $\kappa_T = 3 \times 10^{-6} - 7 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ in the deep-ocean (Kelley, 1984)).
B. Light attenuation technique

Variations in transmitted light were measured with a video camera and two light sheets set up on both sides of the tank, following the same approach as Carazzo and Jellinek (2013). The images obtained in the RGB system were converted into grayscale images, and the intensity of each pixel was determined using ImageJ (Supplementary Fig. 2). The calibration law giving the light attenuation \( \frac{I}{S} \) received by the camera at a given location along the cloud as a function of the mean particle mass fraction \( C \) is of the form

\[
\log \left( \frac{I}{S} \right) = -aC,
\]

where \( I \) is the intensity of the transmitted light, \( S \) is the intensity of the incident light, and \( a \) is a constant parameter that depends on the absorption properties of the fluid and particles, and on the distance traveled by the light through the particle-fluid mixture. This relationship is found to capture the behavior of our experimental materials in a simplified 2D Hele-Shaw geometry in the limit of dilute suspension \( (5 \times 10^{-4} < C < 5 \times 10^{-2}) \) such as \( a = 1.2 \) with \( R^2 = 0.953 \). Moreover, it is fully consistent with previous experimental results on particle sedimentation in dilute suspensions (Bergougnoux et al., 2003; Chehata Gomez et al., 2008, 2009).

Estimating the value of \( a \) is not straightforward in our experimental configuration because we cannot easily impose \( C \) in the cloud (which differs from the particle content imposed at the source of the vertical plume). However, rewriting Eq. (B.1) to express the ratio of the particle concentration in the cloud to its initial value \( C_0 \) as a function of \( I \) gives

\[
\frac{C(t)}{C_0} = \frac{\log \left( \frac{I(t)}{I_0} \right)}{\log \left( \frac{S}{S_0} \right)} + 1,
\]

where \( I_0 \) is the intensity of the transmitted light immediately after the cloud emplacement \( (t = 0) \). Eq. (B.2) shows that the gray level averaged intensity of all the pixels of the region of interest at \( t = 0 \) (i.e., when \( C = C_0 \)) can be used to estimate \( I_0 \) (stage t2 in Supplementary Fig. 2), whereas the averaged gray-scale intensities after the sedimentation is complete (i.e., when \( C = 0 \)) can be used to estimate \( S \) (stage t4 in Supplementary Fig. 2). By means of this calibration, Eq. (B.2) can be used to infer the dimensionless particle concentration, \( \frac{C(t)}{C_0} \), from the amount of light attenuation by particles, \( I(t) \). As discussed in Bergougnoux et al. (2003), the light attenuation is mainly due to light scattering by the surfaces of the particles because the index of refraction of the particles differs from that of the fluid. The effect of multiple scattering is considered to be small (within the error bars in Fig. 4 of the paper) at the low concentration used in this study, which is consistent with our 2D experiments and previous published work (Bergougnoux et al., 2003; Chehata Gomez et al., 2008, 2009).
Supplementary Figure 2: Representation of the method used to measure the rate of change of particle content relative to the initial concentration (exp. 10). (A) After conversion into grayscale (horizontal and vertical field widths, 10 cm). (B) Corresponding surface plot obtained using ImageJ representing the intensity of each pixel in panel A (0 = black; 1 = white). Stage t1: prior to the experiment; Stage t2: immediately after the cloud emplacement. This stage corresponds to the initial condition \( t = 0 \) min prior to particle sedimentation \( (I = I_0) \). The dashed line corresponds to the cloud/salt water layer interface; Stage t3: during the vertical transfer of particles from the cloud to the lower layer of salt water \( (t = 10 \) min). Stage t4: after the complete transfer of particles from the cloud to the lower layer of salt water \( (t = 150 \) min). Some particles are held in suspension in the lower layer by convection but all the particles left the cloud.
C. Dissolution model

The model assumes that dissolution is rate-limited by the diffusion of solute across relatively thin compositional boundary layers (CBL) around each particle (Kerr, 1994, 1995). The dissolution rate of a given particle is given approximately by:

\[ \frac{dm}{dt} = A \Delta c, \]

where \( m \) is the mass of dissolving substance, \( A \) is the particle surface area, \( \Delta c \sim 1 \) is the effective concentration difference (i.e., thermodynamic disequilibrium) driving dissolution, and \( K \) is an effective mass transfer coefficient. This coefficient is a well-characterized function of the flow regime governing the CBL thickness, which depends on the particle Reynolds number (\( Re_p \)) and the ratio of the diffusivities of momentum and solute (\( \nu/\kappa_s \)) (Kerr, 1995; Nield and Bejan, 2006), and a more complicated function of the local pH-Eq conditions in the CBL (Sato, 1960; Luther, 1987; Moses and Herman, 1991). The dissolution experiments of Feely et al. (1987) give \( K = 8 \times 10^{-10} \text{kg m}^{-2} \text{s}^{-1} \) for pyrite particles held in suspension in deep-ocean waters.

The area available for dissolution can be determined from the total grain size distribution (TGSD). Assuming that the particle-fluid mixture is dilute (Feely et al., 1992) and taking a spherical geometry for the particles, we write

\[ A = \sum_{i=1}^{n} N_p(i) A_p(i) = \sum_{i=1}^{n} N_p(i) \pi d_p^2(i), \]

where \( n \) is the maximum particle size remaining in the cloud, \( N_p(i) \) is the number of \( i \)-sized particles, \( A_p(i) \) is the surface area of a single \( i \)-sized particle. For each class of \( i \)-sized particles, the number of particles is given by the ratio,

\[ N_p(i) = \frac{V_1(i)}{V_p(i)} = \frac{6 C_1(i)}{\rho_p \pi d_p^3(i)} = \frac{6 y_1(i) C_1}{\rho_p \pi d_p^3(i)} \]

where \( V_1(i), C_1(i), \) and \( y_1(i) \) are the volume, concentration, and mass fraction of \( i \)-sized particles in the cloud, respectively, and \( V_p(i) \) is the volume of a single \( i \)-sized particle. Replacing \( N_p(i) \) by its expression gives a general expression for the surface area available for dissolution,

\[ A = \frac{6 C_1}{\rho_p} \sum_{i=1}^{n} y_1(i) \frac{d_p(i)}{\pi d_p^2(i)}. \]

However, rigorous constraints on the TGSD in hydrothermal clouds are not available (e.g., German and Von Damm, 2004). Thus, we only consider pyrite particles with a grain size distribution narrowly centered on a single value (\( d_p \)) decreasing with time at a rate of \( 5.3 \times 10^{-12} \text{cm s}^{-1} \) (Feely et al., 1987). Bearing in mind this assumption, we write

\[ A = \frac{6 C_1 y_1}{\rho_p d_p}, \]

where \( C_1 \) is in units of mass/volume and \( A \) is in units of surface area/volume. For pyrite, we assume that \( y_1 = 0.1 \) based on recent field measurements within hydrothermal clouds (Toner et al., 2012) and we take \( \rho_p = 5,000 \text{kg m}^{-3} \).
References


