

TITLE: Diffusive and ebullitive transport of methane and nitrous oxide from streams: Are bubble-mediated fluxes important?

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ABSTRACT:

Journal of Geophysical Research: Biogeosciences Volume 116, Issue G4 Free Access Diffusive and ebullitive transport of methane and nitrous oxide from streams: Are bubble-mediated fluxes important? Helen M. Baulch, Helen M. Baulch Environmental and Life Sciences Graduate Program, Trent University, Peterborough Ontario, Canada Now at the School of Environment and Sustainability and the Global Institute for Water Security, National Hydrology Research Centre, University of Saskatchewan, Saskatoon, Saskatchewan, Canada Search for more papers by this author Peter J. Dillon, Peter J. Dillon Department of Environmental and Resource Studies, Trent University, Peterborough, Ontario, Canada Search for more papers by this author Roxane Maranger, Roxane Maranger Department of Biological Sciences, University of Montreal, Montreal, Quebec, Canada Search for more papers by this author Sherry L. Schiff, Sherry L. Schiff Department of Earth and Environmental Sciences, University of Waterloo, Waterloo, Ontario, Canada Search for more papers by this author Helen M. Baulch, Helen M. Baulch Environmental and Life Sciences Graduate Program, Trent University, Peterborough Ontario, Canada Now at the School of Environment and Sustainability and the Global Institute for Water Security, National Hydrology Research Centre, University of Saskatchewan, Saskatoon, Saskatchewan, Canada Search for more papers by this author Peter J. Dillon, Peter J. Dillon Department of Environmental and Resource Studies, Trent University, Peterborough, Ontario, Canada Search for more papers by this author Roxane Maranger, Roxane Maranger Department of Biological Sciences, University of Montreal, Montreal, Quebec, Canada Search for more papers by this author Sherry L. Schiff, Sherry L. Schiff Department of Earth and Environmental Sciences, University of Waterloo, Waterloo, Ontario, Canada Search for more papers by this author

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Abstract [1] Using measured rates of bubble release and diffusive gas transport (also termed surface aeration), we address the role of these transport mechanisms in emissions of nitrous oxide and methane from four streams. While ebullition in streams and rivers has received little study, we found that ebullition was an important mode of methane emissions, contributing 20%–67% of methane emissions (among streams). Nitrous oxide emissions via ebullition were negligible (<0.1% of diffusive emissions). Total methane emissions (ebullition + diffusive transport) were over ten times greater than N₂O emissions in terms of CO₂ equivalents. Rates of bubble release were highly variable, ranging from 20 mL m⁻² d⁻¹ to 170 mL m⁻² d⁻¹ (seasonal average among streams, with volumes reflecting ambient temperature and pressure). Methane was the most abundant of the bubble gases that were measured (26% by volume on average among streams), followed by carbon dioxide (1% on average) then nitrous oxide. Average bubble nitrous oxide concentrations were below atmospheric mixing ratios for the majority of streams; however, one stream showed concentrations as high as 3600 ppbv. Sediment characteristics were strong predictors of bubble composition. Concentrations of methane and nitrous oxide were positively related to the proportion of fine sediments. High methane concentrations in bubbles were related to high sediment organic carbon.

Key Points

- Ebullition is an important mode of methane emissions from streams
- Bubble gas concentrations and emissions are related to sediment characteristics
- Methane emissions significantly exceed nitrous oxide emissions in terms of GWP

1. Introduction

Aquatic Ecosystems as Sources of Greenhouse Gases [2] On a global basis, lakes, reservoirs and wetlands have a significant role in global greenhouse gas balances. Lake emissions have been estimated at 70–150 Tg y⁻¹ of carbon in the form of carbon dioxide (CO₂-C) [Cole et al., 2007] although a recent reassessment places this number as high as 530 Tg CO₂-C y⁻¹ [Tranvik et al., 2009]. Methane (CH₄) emissions from lakes are on the order of 8–48 Tg y⁻¹ [Bastviken et al., 2004], but this number may also be an underestimate [Walter et al., 2006]. Wetland CH₄ emissions are globally more significant than lakes at between 145 and 231 Tg of CH₄ per year [Denman et al., 2007]; however, many wetlands appear to be net sinks of CO₂ [Bridgman et al., 2006]. Reservoirs are important not only for their large emissions, but also because they are considered a form of land-use change, and are factored into anthropogenic

greenhouse gas budgets. Emissions from reservoirs are estimated at 280 Tg of CO₂-C [Cole et al., 2007], and 70 Tg of CH₄ per year globally [St. Louis et al., 2000]. [3] Fluvial systems have received less study in terms of their role in CH₄ emissions [Bastviken et al., 2011]; however, existing data suggest that they function as CH₄ sources to the atmosphere [Jones and Mulholland, 1998]. The greenhouse gas receiving significant study in fluvial ecosystems is nitrous oxide (N₂O), due to an early model suggesting that agricultural and sewage N inputs into streams, rivers and estuaries could lead to 20% of the world's anthropogenic N₂O emissions [Seitzinger and Kroeze, 1998]. Based on accumulating evidence that river and stream emissions are overestimated, the IPCC has recently downscaled N₂O emissions estimates from fluvial ecosystems [Eggleston et al., 2006]. However, new data suggests IPCC methods may now significantly underestimate riverine N₂O fluxes [Beaulieu et al., 2011]. Carbon dioxide emissions from rivers are estimated as 230 Tg CO₂-C per year [Cole et al., 2007]. Estimates of stream fluxes (based on stream heterotrophy, which likely represent a lower bound for emissions) suggest that stream emissions are greater than river emissions at 320 Tg CO₂-C y⁻¹ [Battin et al., 2008]. These high fluxes are supported by measurements from boreal streams where streams function as strong CO₂ sources in the landscape [Teodoru et al., 2009]. In sum, inland waters may emit as much as 1400 Tg CO₂-C per year [Tranvik et al., 2009].

Production and Emission of Greenhouse Gases [4] Numerous pathways contribute to greenhouse gas production. Classical denitrification can lead to N₂O production, as do the pathways nitrification and nitrifier-denitrification. The two dominant CH₄ production pathways are acetate fermentation (aceticlastic methanogenesis) and H₂-dependent (hydrogenotrophic) methanogenesis. Aerobic respiration, denitrification, aceticlastic methanogenesis and other pathways contribute to CO₂ production. [5] Gases are emitted from aquatic ecosystems by three major mechanisms. The first and most frequently studied is diffusive transport (also referred to as air-water gas exchange or surface aeration). However, plant and bubble-mediated (ebullitive) transport can be important, particularly for CH₄ [Dacey and Klug, 1979; Walter et al., 2006]. Gas bubbles can be formed in sediments when the partial pressure of the gases exceeds the sum of the pressure on the sediment and the surface tension of water. Hence, bubble production depends on rates of gas production as well as water temperature, water depth, and barometric pressure. Bubble emissions tend to be episodic, triggered by shear stress [Joyce and Jewell, 2003], lowering of the water table [Chanton et al., 1989], or periods of low atmospheric pressure [Mattson and Likens, 1990]. [6] A major limitation of our understanding of the importance of streams and rivers in greenhouse gas emissions is in our focus on diffusive gas transport across the air-water interface. The importance of bubble-mediated emissions, especially of N₂O, has largely been overlooked with the assumption that diffusive transport is the dominant emission mechanism. We assess this assumption in a series of streams in southern Ontario, Canada by contrasting rates of gas transport via these two mechanisms. We also assess rates of CH₄ emissions and the role of ebullition in CH₄ transport ? a topic that has received relatively little study in small streams. Finally, we identify predictors of gas concentrations and fluxes among streams.

2. Site Description [7]

We studied four 2nd-5th order (Strahler) streams in southern Ontario, Canada. These are north temperate, hard water streams, with significant agricultural land use in their catchments. Monoculture (continuous cultivation of annual crops) constitutes 13%-22% of land area, mixed cropping (rotations of annual and forage crops) constitutes 10%-31% of land area, rural land uses (forage crops, idle lands, hay, pasture and marginal lands) include 12%-44% of catchment areas, and wetlands constitute 9%-25% of land area. Urban areas are <3% of catchment areas in all streams [Ontario Ministry of Natural Resources, 2006]. The highest proportion of wetland area is observed in Layton Creek (25% of catchment) and Jackson Creek (21% of catchment), while the greatest proportion of wooded area is in the Black River (24%) and lowest proportion in Mariposa Brook (9%). Mariposa Brook has the greatest proportion of land in agricultural land use (monoculture, mixed or rural). We sampled two sites (separated by 3.9-8.2 km; Table 1) in each stream. Land use data reflect the most downstream site sampled [Baulch, 2009]. Study streams and sampling sites were selected to represent small streams of this region, while ensuring easy access, and minimizing the number of inflows between sites. All streams were shallow (<50 cm mean depth), with relatively low slope (0.25 to 1.43°; Table 1). A map of study sites, and additional site characterization data are available elsewhere [Baulch et al., 2011a].

Location	Site Coordinates	K Measurements	Stream Characteristics	K600 (day ⁻¹) at 20°C	Method	Discharge (L s ⁻¹)
Black River Upstream	N44° 09.476, W079° 21.673	5.4, 4.9	SF6, O2 isotope	98, 62	98 (39)	0.50, 8.2
Black River Downstream	N44° 11.815, W079° 20.150	(from above) 87 (32)				
Jackson Creek Upstream	N44° 15.976, W078° 28.383	5.3, 7.8	SF6	43, 60	76 (14)	1.43, 6.6
Jackson Creek Downstream	N44° 16.799, W078° 24.850	3.3, 2.5	SF6	120, 145	282 (231)	
Layton River Upstream	N44° 16.071, W078° 59.075	10.6, 11.4	SF6, O2 isotope	3, 6	30 (17)	0.25, 3.9
Layton River Downstream	N44° 14.501, W078° 58.876					
Mariposa Brook Upstream	N44° 23.148, W078° 52.545	1.5	SF6	2	2 (0)	1.03, 7.0
Mariposa Brook Downstream	N44° 20.915, W078° 52.084	3.3	O2 isotope	13	24 (9)	

3. Materials and Methods [8]

At each site, we measured stream

discharge and water chemistry, the rate of bubble release, bubble gas concentrations, and the rate of gas emissions driven by diffusive transport (determined using measurement of piston velocity and dissolved gas concentrations). Sediment analyses were also performed at each site. Water Chemistry, Stream Discharge, Temperature, Pressure [9] Water samples were obtained in 750 mL PET bottles from midstream. A total organic carbon analyzer (Shimadzu TOC-V analyzer) was used to analyze concentrations of dissolved organic carbon (GFF filtered). pH was measured using electrodes (Mantech PC titrator; Mantech Inc, Guelph Canada). Nitrogen species were GFF filtered then analyzed colorimetrically. Nitrate was analyzed using the red azo dye method, following reduction to nitrite, then corrected for measured nitrite [Ministry of the Environment, 2001]. Ammonium was analyzed using the phenate-hypochlorite method [Ministry of the Environment, 2001]. Samples for total phosphorus analysis were obtained directly in glass vials, then analyzed using the ammonium-molybdate-stannous chloride method [Ministry of the Environment, 1994]. [10] Depth, velocity, and discharge were measured at each site using a Swoffer 2100 velocity meter and applying the velocity-area method. Air and water temperatures were measured using a Fisherbrand Traceable Thermometer. Barometric pressure was measured using a handheld barometer (Kestrel). Rate of Bubble Release [11] We deployed transparent inverted-funnel style bubble traps [Molongoski and Klug, 1980] from late May to 9 October 2007 to determine the volume of bubbles emitted from the benthic substrate. The initial sampling design involved only three traps being installed at each site. After the first sampling, we installed additional traps (bringing totals to 5-10 traps per site), to account for observed high spatial variability. Our sampling design (including the number of streams, sites, and traps per site) represents a tradeoff between more spatially intensive sampling, resulting in more constrained emissions for a single stream, and more extensive sampling across streams. [12] Bubble traps had a bottom area of 530 cm² which narrowed into an upper trap portion with a diameter of 2.2 cm. Traps were affixed to a galvanized metal pole (which was inserted into the substrate) and installed 10-30 cm below the water surface and at least 10 cm above the benthic substrate, allowing water flow below the trap. The minimum water depth which we could install bubble traps was 25 cm. The depth of traps at the end of the sampling period is reported in Table 2. Data for the first two weeks following trap installation was omitted to limit effects of initial sediment disturbance on ebullition rates. Although the use of this apparatus in flowing water may also contribute to sediment disturbance, very slow stream velocities during the study period (Table 2) suggest these effects were small. Slow water flow also ensured bubbles emitted from below the trap were transported upwards into the vessel (rather than moving downstream with water flow). Table 2. Mean Stream Velocity, Sediment Characteristics and Water Depth at Bubble Trap Locations at the End of Sampling Period

Location	Site	Bubble Traps	Sediment Characteristics	Sediment Size Fractions (% by Weight)	Mean Stream Velocity (m s ⁻¹)	Mean Water Depth (cm)
Black River Upstream	0.07	38 (5)	8	2.1 (0.9)	69.6 (3.6)	2.1 (0.8)
Black River Downstream	0.07	56 (3)	6	1.4 (0.6)	63.7 (3.7)	2.3 (0.9)
Jackson Creek Upstream	0.08	41 (1)	8	2.5 (0.7)	50.7 (4.6)	5.9 (1.6)
Jackson Creek Downstream	0.06	59 (3)	9	1.7 (0.6)	78.7 (3.9)	17.2 (4.5)
Layton River Upstream	0.02	56 (3)	8	18.6 (2.0)	24.3 (4.4)	8.4 (3.3)
Layton River Downstream	0.03	45 (2)	9	14.1 (1.3)	26.5 (2.8)	8.3 (4.1)
Mariposa Brook Upstream	0.01	65 (4)	10	2.5 (0.2)	46.4 (3.1)	1.1 (0.3)
Mariposa Brook Downstream	0.01	42 (2)	6	2.4 (0.3)	69.9 (4.3)	17.3 (2.4)

a Mean values are indicated with standard error in parentheses. b During the period for which emissions via ebullition and diffusive transport are contrasted (11 June-11 October 2007). [13] Traps were filled with water (no headspace) at the start of each sampling period. Gases emitted from below the trap surface accumulated in the upper (narrow) portion of the funnel and were sampled to determine bubble volume using a luer-lock fitting at the top of the trap attached to a 60 mL syringe (approximately every 2 weeks). Where very high emissions occurred, an inverted water-filled 1000 mL graduated cylinder was used to determine the volume of bubbles emitted. Care was taken to avoid disturbing the benthic substrate when sampling the traps, including maintaining a minimum (lateral) distance of 1 m from the traps, and having one person continuously monitor the traps to ensure no bubble plumes were released from the bottom substrate as a consequence of our sampling. When there was evidence of muskrats tampering with the traps, or evidence that sampling induced substrate disturbance (affecting the volume of gas collected), affected data were excluded. This resulted in the exclusion of 4% of bubble flux measurements. Gas Sampling [14] Water samples were obtained for dissolved gas analysis from midchannel, approximately 20 cm below the water surface. Sampling was performed at each study site on 9-10 occasions (between 15 May to 30 October 2007). Samples were taken in 125 mL borosilicate glass serum bottles with no headspace, capped with pre-baked butyl rubber stoppers and field preserved with saturated mercuric chloride (0.4% final v/v). [15] Following sampling for dissolved gases, trapped bubble volume and water chemistry, we obtained bubble samples from the benthic substrate

for analysis of bubble gas concentrations. Samples were obtained by walking transects of the stream channel (perpendicular to streamflow at each site) and periodically disturbing the benthic substrate with a funnel attached to the body of a 60 mL syringe and luer-transfer port. Multiple transects were walked until at least 30 mL of gas was obtained, which was then transferred into a pre-evacuated vial. This process was then repeated 3 times to obtain triplicate samples which were analyzed using gas chromatography (GC). Bubble gas sampling was performed on 9 or 10 dates at each study site (see results for exact timing). [16] We opted to analyze fresh bubbles from the sediments rather than those trapped within the headspace of bubble traps because bubbles within the traps will undergo equilibration with stream water. However, fresh bubbles may have undergone further equilibration with surrounding pore waters prior to natural emission. In addition, bubbles undergo equilibration as they travel through the water column to the surface. Because the length of the apparatus used in fresh bubble sampling (~40 cm) was sometimes less than the stream depth (Table 2), the extent of equilibration may have differed between naturally emitted samples and the values we report. This effect is most important for highly soluble or reactive gases such as H₂S and CO₂. In contrast, effects on N₂O and CH₄ are expected to be minimal [Chanton and Whiting, 1995]. However, our CO₂ measurements could represent a slight overestimate of the CO₂ concentration the same bubbles would attain at the water-air interface. Gas Analyses [17] Dissolved gas samples were analyzed using a Varian CP-3800 gas chromatograph following headspace equilibration. CH₄ was analyzed by FID following separation using a Haysep D column (80/100 mesh, 0.5 m, 1/8" stainless steel). N₂O was analyzed by ECD using the same column. Equilibrium concentrations for dissolved gases were calculated using measured temperature and barometric pressure [Yamamoto et al., 1976; Weiss and Price, 1980], assuming atmospheric concentrations of 320 ppbv N₂O and 1.85 ppmv CH₄. Given the long atmospheric residence time of N₂O, it is expected to be well mixed in the atmosphere [Stein and Yung, 2003], which supports our assumption of atmospheric mixing ratios. However, regional variation in CH₄ concentrations is likely, and our assumption of constant atmospheric concentrations may bias our flux estimates. Where dissolved CH₄ is highly supersaturated, this will have only small effects on CH₄ flux estimates (i.e., see equation (1)). [18] All samples were obtained during the daytime. This may result in a slight underestimate of diffusive CH₄ fluxes. Diel sampling of these streams indicated that sampling near solar noon underestimated diffusive CH₄ fluxes by 9% in these streams, when compared to time-weighted samples over a 24-h sampling period [Baulch, 2009]. Daytime sampling underestimates diffusive N₂O flux from these streams by ~5% [Baulch, 2009]. Lateral variation in dissolved gas concentrations across the stream channel was observed (average of 6% for N₂O, 11% for CH₄), but systematic differences between samples obtained from the middle of the stream channel and samples obtained near stream banks were not observed. [19] Bubble samples were analyzed on two instruments. Approximately 60% of samples were analyzed on the Varian gas chromatograph as described previously. The remainder of samples were analyzed using a Shimadzu GC-2014 with a Tekmar 7050 autosampler and 9 mL vials with thick butyl rubber septa. Gases were separated on a Poropak Q column (80/100 mesh size). Samples for CH₄ analyses were diluted with UHP N₂ or helium. Duplicate bubble samples run on both gas chromatographs showed agreement within 3% (CH₄), 2% (CO₂), and 7% (N₂O) between instruments. Gas Emissions via Ebullition [20] The masses of N₂O, CO₂ and CH₄ released by ebullition were calculated by determining the number of moles of total bubble gas released (using measured volume, air temperature and atmospheric pressure), and multiplying this value by measured gas concentrations in the fresh bubbles (at the start and end of the sampling period) for each site. Average, time-weighted emissions were then calculated, accounting for the duration of each sampling interval. Error in ebullitive flux estimates was estimated using Monte Carlo uncertainty analysis [Beck, 1987]. This method allows us to constrain uncertainty associated with both the volume of bubbles, and bubble gas concentrations. The volume of gases emitted and the concentration of gases were represented as a normal distribution using measured means and standard deviations, and truncating distributions to omit negative values [Decisioneering, Inc., 2001]. Ebullitive emissions of both N₂O and CH₄ were estimated by subsampling these distributions 1000 times using stratified random (Latin Hypercube) sampling in the software package Crystal Ball 2000.2 (Decisioneering, Inc., Denver, Colo.), resulting in a probability distribution for ebullitive gas fluxes. Gas Emissions via Diffusive Transport [21] Gas fluxes due to diffusive transport were estimated using the two-layer model of diffusive gas exchange [Liss and Slater, 1974]: where k is the piston velocity (m h^{-1}). The piston velocity is sometimes termed the gas transfer velocity, and is equal to the gas transfer coefficient K times the mean depth; C_S is saturation concentration (mol m^{-3}); C_L is the measured concentration (mol m^{-3}). [22] In this model, there are two interfacial layers, one in the water at the water surface, and one in air, immediately overlying the water surface. For CH₄ and N₂O, air-phase resistance is negligible [Liss and Slater, 1974], and the main resistance to gas transfer across the air-water interface is molecular diffusion through the surface layer of the water. The depth of this surface layer, which dictates the length of the diffusion pathway, is related to turbulence. Within shallow fluvial ecosystems, benthic turbulence is the primary process affecting rates of gas exchange [Raymond and Cole, 2001]. [23] Our measurements of rates of air-water gas transfer (Table 1) are described in detail elsewhere [Baulch et al., 2011a].

Briefly, we measured rates of air-water gas transfer via addition of a gas tracer (sulphur hexafluoride; analysis by GC-ECD as for N₂O) and a conservative tracer at six of our eight study sites [Kilpatrick et al., 1987; Baulch, 2009]. We also used diel O₂ dynamics to constrain the piston velocity [Venkiteswaran et al., 2007]. This method uses an O₂-mass balance model, iteratively fit using constrained range of possible rates for respiration (0–5000 mg O₂ m⁻² h⁻¹), photosynthesis (0–5000 mg O₂ m⁻² h⁻¹) and piston velocity (0.01 to 0.50 m h⁻¹) [Venkiteswaran et al., 2007]. The model was run with different parameter combinations (i.e., rates of respiration, photosynthesis and piston velocity) to obtain the lowest sums of squares between measured and modeled data. All model runs where an r² of >0.8 were averaged to obtain a mean estimated piston velocity. [24] In one stream (Black River) where substrate, depth, and flow varied little between sites (mean differences: proportion substrate >0.25 mm, 11%; mean depth 16%; mean discharge 9%), we used upstream gas transfer measurements to estimate gas transfer at our downstream site. At all other sites, one or two direct measurements of gas transfer were made at each study site (Table 1). Measured rates of gas transfer were converted to the stream temperature [Thomann and Mueller, 1987] and gas of interest (from SF₆ or O₂ to CH₄ or N₂O) using Schmidt number scaling [Wanninkhof, 1992]. We assumed a Schmidt number exponent of 2/3 which reflects smooth surfaces (in contrast to rough surfaces, where a Schmidt number exponent of 1/2 is typically assumed [Jähne et al., 1987]). We report rates of air-water gas transfer in customary units, as the gas transfer coefficient for CO₂ at 20°C (K₆₀₀; Table 1). [25] Depth, velocity, slope and substrate type are important factors affecting benthic turbulence, and depth and velocity can vary over time (Table 1). To account for this temporal variation which is likely to affect rates of gas transfer, we use two approaches. First, in 5 of 8 study sites we use two measurements of gas transfer coefficient (Table 1) and report the range of estimates resulting from these measured values (in the remaining 3 sites, only a single measurement was made). This assumes that gas transfer coefficient was constant through the study period. Second, we applied a model-based approach to extrapolate measured values to different flow conditions (varying depth and velocity) [Moog and Jirka, 1998]. We calibrated three empirical models [O'Connor and Dobbins, 1958; Bennett and Rathburn, 1972; Owens et al., 1964] to individual sites, using the ratio of measured K to modeled K under the flow conditions when K was determined [see equation (2); Moog and Jirka, 1998], then applied these calibrated empirical models to predict gas transfer coefficient, using measurements of stream depth and velocity for each sampling occasion. The use of three models is intended to reflect error associated with model assumptions. where K_{PC} is the calibrated gas transfer coefficient, K_m is the measured gas transfer coefficient, K_P is the predicted gas transfer coefficient under measurement conditions, and n reflects the number of measurements. This equation is presented in its original form, which uses the gas transfer coefficient, a value equal to the piston velocity (k) divided by depth. [26] We used K_{PC} values to determine whether dissolved gases at upstream sampling sites were likely to affect gas composition at the downstream location. In all cases, the distance between sampling sites was great enough that dissolved gases at upstream sites had minimal effects (<5%) on concentrations at downstream sites based on first-order loss equations [Chapra and Di Toro, 1991]: where Dist_{5%} is the distance at which 5% of a gas will remain in solution, v is stream velocity and K_{PC} is the calibrated gas transfer coefficient. [27] Dissolved gas concentrations and bubble gas concentrations are reported for the period 15 May to 30 October 2007. Emissions via diffusive transport and ebullition are calculated from 11 June to 11 October 2007 for the Black River, Jackson Creek and Layton Creek. However, in Mariposa Brook, where changes in discharge led to changes in piston velocity and added considerable uncertainty to our diffusive transport estimates, we restrict the time series to 3 July to 12 September 2007 (upstream site) and 11 June to 12 September 2007 (downstream site). Mean (time-weighted) diffusive flux estimates for the study period were determined using mean fluxes for each sampling interval (mean of start and end of each period), then multiplying by the duration of that interval, and dividing by the length of the study period. Sediment Analyses [28] Duplicate sediment cores were obtained from under each bubble trap at the end of the study (October 2007). A PVC core tube (10 cm diameter, 7 cm depth) was pressed into the stream substrate. The base was sealed from below using a plastic sheet or hand, then the core tube was removed, and its contents were transferred to pre-labeled bags. At two sites (Mariposa Brook, downstream site, Jackson Creek, downstream site), we could not obtain cores from under all traps, because the rocky substrate prevented use of our core tube. Samples were split to allow analysis of dry weight percentage (comparing wet weight to weight following drying at 60°C) and sediment size fractions. Samples for size fractionation were wet sieved following treatment with a dispersant to prevent clumping (sodium hexametaphosphate) [Pope et al., 2000]. Sample retained on the largest sieve (2 mm) was discarded. Material passing through the smallest sieve (0.0625 mm) was poured into evaporating vessels, dried, weighed, and corrected f

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