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Evolution and relationship between 3 dissolved gases (oxygen, methane, and carbon dioxide) over a 10-year period (1994-2003) in a river downstream of a new intertropical dam

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Introduction

The emission to the atmosphere of CO2 and CH4 produced by large artificial reservoirs created in tropical forests is a subject of scientific discussions (Delmas et al 2001 and SIL sessions in 2001-2004). The dynamics of the dissolved part of these gases released into the downstream river has been much less studied. For Petit Saut (PS) hydroelectric reservoir (French Guyana), the significant contribution of dissolved methane (DM) oxidation to the dissolved oxygen (DO) consumption in the downstream Sinnamary river (260 m³ s⁻¹ mean annual discharge) was shown (Gosse 1994, Gosse et al 2000). The installation in February 1995 of a two-falls aerating weir (described in Gosse et al 1997) in the plant outlet canal became necessary to exchange O2 and CH4 with the atmosphere in order to both turbine the planned discharges and guarantee the minimum regulatory DO concentration of 2 mg/L in the downstream Sinnamary. The DO and DM budgets in the 40 km river stretch downstream of the dam were previously examined for the 1994-1998 period (Gosse et al 2000, Gosse et al. 2002). In this paper, we actualise theses budgets considering the 1999-2003 data set and we add an analysis of dissolved CO2 (DC). The main remarkable change is the significant decrease in the velocity of methane oxidation by comparison to the previous years. We calculate that in 2001 (before the lowering of the upper fall) and in 2003 (after its complete lowering) no more than half of the DM released by the weir in its present lowered configuration was oxidised along the stretch in the usual range of daily turbined discharges, and that less than 2/3 of the DO sink appearing along the stretch was due to methane oxidation. It is assessed that the internal production of CO2 from oxidation processes along the stretch was replacing around half of the CO2 flux emitted by aeration from the stretch to the atmosphere in 2003. The methods used for gas measurements are specified in Galy-Lacaux (1996), Delmas et al (2004), and Richard et al (2004), where figures showing longitudinal profiles of the dissolved gases can also be found.

Experimental site

Filling of PS reservoir began in January 1994. Due to the efficiency of the weir, the functioning of the hydroelectric plant has been continuous since March 1995. Since the first filling of the reservoir in June 1995, the volume of the reservoir varies between 2.3 and 3.5 km³ depending on the alternation from wet to dry season. Except for high floods during which the bottom or surface gates need to be opened, or for prolonged dry seasons which require to turbine less, the daily discharges released by the dam are totally turbined and are generally in the 150-270 m³ s⁻¹ range, with an elimination at the weir -in its initial configuration - of around 80% of the DM contained in the flow entering the turbines. For theses usual turbined discharges, the 40 km stretch of the river under study - starting at the outlet of the weir which is 0.1km downstream of the dam - can be roughly divided in 2 rather equal stretches; a upper stretch - close to 100 m wide between the densely vegetated banks - where wind is very low and tide action has a low effect on water depth and flow direction; and a lower stretch ending at Pointe Combi (PC), which is around 20% wider, under the influence of the coastal winds, and significantly affected by tides which generate a 12 hours cycle of water depth (with an amplitude in the order of 1 metre at PC). No salinisation has been observed however, a distance of 25 km separating PC from the sea. To understand DM and DO budgets in the stretch of river under study, the mean velocity (U in m/s) and mean depth (H in metres) of the running flow between the weir and PC can be roughly schematised in the 100-250 m³ s⁻¹ range by the following equations which filter tide action and neglect the rather stagnant areas situated on the banks, under the dense riparian vegetation: H=1.56 Q $^{0.193}$ and U= 0.007 Q $^{0.75}$ with Q discharge crossing the dam in m³ s⁻¹ (Gosse et al 2002). As higher margins above the downstream DO threshold of 2 mg L⁻¹ were progressively observed (see below), the upper fall of the weir has been practically totally suppressed (a small swirl is still present) to maximise the power production, with an identical abstraction of 1.2m in December 2001 and March 2003.

Dissolved methane budget

Due to the high air-water mixing taking place at the outlet of the bottom gates which delivered a reserved flow of 100 m3/s, there were - with the exclusion of turbining tests - low concentrations of DM in the water released by the dam from 1994 to February 1995, with a concentration lower than 0.2 mg/L before June 1994 and a maximum of 0.5 mg/L reached in January- February 1995 (Gosse et al 2000, Gosse et al 2002). In relation to the decrease of the DM stock in the reservoir, the DM concentrations measured in the turbines entrance have progressively decreased from 7.5 mg/L in 1995 to 3 mg/L in 2003 on annual average, with a yearly cycling giving lower and higher concentrations before and at the end of the dry season respectively). The mean concentration over the March 1995-December 2002 has been assessed at 4.7 mg L⁻¹(Richard et al, 2004). Despite the aeration effect of the weir, the monthly concentrations of DM measured in the Sinnamary in the near downstream of the aerating weir have been quite significant compared to classical rivers with a mean annual value (neglecting short floods events needing the opening of dam gates) of 1.5 mg/L

in 1995, 1.4 mg/L in 1996, 1.1 mg/L over the years 1997-1998, 0.7 mg/L and 0.55 mg/L over the years 2000-2001. It has increased after 2001 due to the lowering of the upper fall of the weir and a resulting loss in aeration efficiency, with a mean value of 0.85 mg/L over the January 2001-February 2003 period corresponding the first lowering, and 1.1 mg/L over the last 10 months of 2003 corresponding to the 2nd lowering. The data tend to indicate that between 70 and 75% of the DM has been eliminated by the weir after its first lowering, and between 60% and 65% after the second one. As regards the longitudinal evolution of DM concentration downstream, it was shown that until 1998 - disregarding the short floods events which require the opening of the bottom gates and reduce the aeration efficiency of the weir - there was only traces of DM in the water at PC, with concentrations not exceeding 0.05 mg/L for daily turbined discharges in the 200-270 m3/s range, and lower than 0.01 mg/L under 150 m3/s (Gosse et al, 2002). 2 factors could explain this DM disappearance: aeration at the river surface and DM oxidation by bacteria. The dominance of DM oxidation was shown experimentally, both by in vitro analysis of DO and DM variations in water samples and by in situ measurements of CH4 flux across the air-water interface conducted in 1995 for 2 discharges, using a floating chamber linked to a drifting boat (Galy-Lacaux 1996; Lacaux and Delmas, 1995). At 220 m³ s⁻¹ the measured air emissions of DM were rather well described at the 5 measurement points - distributed between the weir and Crique Venus (CV) at 23 km downstream of the dam - by the equation F = 1.8 DM, with F in g m⁻² day⁻¹ and DM in mg/L, whereas the equation F = 0.5 DM was better suited for the 2 measurements points (1km and 6 km downstream of the dam) studied at 100 m³ s⁻¹. From these data. (Galv-Lacaux et al 1997) assessed that around 20% of the DM remaining in the river at the outlet of the weir was escaping to the atmosphere between the weir and PC, which means that around 80% was consumed by bacterial oxidation. This order of magnitude was confirmed by a simplified DM model of the river - recalled below extrapolating these local air emissions measurements and using the schematic hydraulic model of the river presented

Since end of 1999, the role of aeration in the progressive longitudinal disappearance of DM has increased to the detriment of DM oxidation, for two main reasons. Firstly, the velocity of DM oxidation has decreased significantly after 1999, as for similar turbined discharges and similar (and even lower) DM concentrations at the outlet of the weir, DM concentrations at PC are greater than before 1999. For instance, in 2003 (statistics on 7 profiles after the 2nd lowering of the weir) for daily turbined discharge in the 150-260 m³ s⁻¹ range and a corresponding mean DM concentration of 1.0 mg/L at the outlet of the weir, the mean DM concentration at PC was 0.3 mg/L, whereas it was more than six times lower in 1996 and 1998 for higher DM concentrations downstream of the weir for similar turbined discharges (Gosse et al 2002). Even in 2001 (statistics on 10 profiles before the 1st lowering of the weir), there was a mean DM content of 0.12 mg/L at PC for an upstream content as low as 0.55 mg/L on average. Secondly, the resulting presence of higher DM concentration in the river, especially in the 20 km sub-stretch where aeration is greater due to wind action, tends to increase the proportion of DM elimination by aeration.

The two recent Hydro-Québec - French campaigns of GHG fluxes measurements conducted the river in 2003 using floating chambers flexibly coupled to a drifting boat bring new data on aeration. In May, for a turbined discharge of 145 m³ s⁻¹, the Quebec and CNRS teams using 2 different equipments (with different integration times) gave similar mean fluxes of CH4 emissions at the 2 upper points studied: 1.35 g m⁻² day⁻¹ around 1.5 km downstream of the dam for a wind of 0.7 m s⁻¹ (all the winds are measured at 1 m above the surface) and 0.77 g m⁻² day⁻¹ at CV for a wind of 0.4 m s⁻¹. Considering the DM measured at these 2 points (1.2 and 0.63 mg/L respectively), the gas transfer velocity k at the air-water interface can be assessed at 1.1 m day⁻¹ and 1.2 m day⁻¹. These values are coherent with the 1995 emission data, as they are inside the 0.5-1.8 m day⁻¹ interval given by the two lower and greater discharges examined in 1995. Only a Quebec measurement is available at PC, the 3rd point of the May campaign in the stretch under study. The emission flux measured was 0.79 g m⁻² day⁻¹ for a DM concentration of 0.26 mg/L and a wind of 2.1 m s⁻¹, which gives a value of k equal to 3 m day⁻¹, which reveals the influence of the penetration of the coastal winds in this area. The December 2003 campaign conducted for a not frequent turbined discharge of 115 m³ s⁻¹ confirmed the presence of higher wind at mi-day in the lower sub-stretch - 1.9 m s⁻¹ at Roche Bravo (RB) 31 km downstream of the dam and 1.5 m s⁻¹ at PC - by comparison to the upper zone -0.4, 0.5 and 0.1 m s⁻¹ at 1.5, 12 and 23 km downstream of the dam. Contrary to the May campaign, the emissions data of 2 teams do not match perfectly in December but they confirm that k tends to be greater downstream than upstream. For the 5 stations, the Quebec team data lead to k values equal to 1.7, 1.4, 2.6, 4.3 and 2.7 m day⁻¹ from the dam to PC, whereas the French team data give 0.5, 1.4, 1.5, 2.9, and 2.3 m day⁻¹. If we consider that the CO2 data (Quebec emission flux + CNRS concentrations in the water) would give a k for CH4 equal to 1.9, 2.4, 2.6, 3.6 and 3.0 at the 5 stations from upstream to downstream (considering a CO2 saturation concentration between 1.4 mg/L and 0.5 mg/L from upstream to downstream and a Schmidt number ratio of 1.025 between the 2 gases at 26.5°C which is the mean temperature of the stretch), it seems that the CO2 data set is more coherent with the Quebec CH4 fluxes. Which tends to indicate that the low value ok k in the upper limit of the stretch given at 100 m³ s⁻¹ by the 1995 emission measurements (and curiously found by the French team in December 2003 as well) underestimate the real situation of aeration in the whole stretch for a comparable level of turbined flow. From this emission data set and the schematic hydraulics of the river presented above, the following equation 1 can be proposed to schematise the aeration coefficient K_2 (day⁻¹) along the stretch under study: $K_2 = (4.14 \text{ U}^{0.5} \text{ H}^{-1.5}) + 0.104 \text{ v}^2$ with v wind velocity (m/s) a 1m. The first term of the equation is the O'Connor and Dobbins formula at 26.5°C (divided by 1.1 which is the Schmidt ratio between O2 and CH4). For a use at a daily scale which filters tide and wind

cycles, it is proposed to take on average - as a first approximation - 0.3 m s^{-1} for v in the first 30 km and no more than 1.5 m s^{-1} for the last 10 downstream km. This limitation is justified by the fact that the gas emissions were measured at mi-day in the downstream part of the river, at a time where the coastal winds are greater, the winds recorded at 10m on the coast at Kourou meteorological station indicating on a yearly scale a mean value of 4.4 m s^{-1} during 9 consecutive hours at mid-day and 1.7 m s^{-1} over the 15 other hours.

A simplified model of the stretch under study was previously proposed: $dDM / dt = -K_2 DM - d_3 DM_0$, with $d_3 (day^{-1}) a$ constant which has to be calibrated and DM₀ (mg/L) the DM concentration at the outlet of the weir (Gosse et al, 2002). The model was calibrated on the 1994-1998 data set, using for K₂ (day⁻¹) the following equation 2 adjusted on the 1995 GHG emission campaign: $K_2 = 0.0023Q-0.1$. The replacement of equation 2 by equation 1 to better fit the 2003 GHG emission campaign doest not change greatly the aeration conditions in the model at the river scale for the usual range of turbined discharges. But it does for discharges around 100 m³ s⁻¹, by doubling the level of K₂, which would need to adapt the schematisation of some processes to better simulate the whole 1994-1998 data set. As regards the present situation, it appears that the model with the new equation 1 represents correctly the longitudinal profile of DM observed during the GHG December 2003 campaign (115 m³ s⁻¹) from the outlet of the weir (1.7 mg/L) to PC 0.3 (mg/L) – including the intermediate values of 0.8 and 0.6 mg/L at CV and RB - by taking $d_3 = 0.28 \text{ day}^{-1}$, which leads to the conclusion that 63% of the DM disappearance along the 40km is due to oxidation and 37% to aeration and that 53% of the DM present at the outlet of the weir is oxidized along the 40 km stretch. These percentages are close - 65%, 35% and 51% respectively - with the GHG May 2003 campaign (145 m³ s⁻¹), for which a good fit of the longitudinal profile of DM observed - from 1.2 mg/L at the outlet of the weir to with 0.63 mg/L at CV and 0.25 mg/L - by taking $d_3 = 0.32 \text{ day}^{-1}$. If we adjust d_3 to reproduce the recent observations of the routine survey in 2003 which shows on average (statistics on 7 profiles) a DM decrease from 1.0 mg/L at the outlet of the weir to 0.3 mg/L at PC in the usual range of turbined discharges 150-270 m³ s⁻¹, we find $d_3 = 0.39$ day⁻¹. The 3 previous percentages become equal to respectively to 65%, 35% and 45%. Which means that 45% of the DM observed at the outlet of the weir in its present lowered configuration is oxidized along the stretch in the mean range of the usual daily turbines discharges. We obtain 52% for year 2001 the last year before the lowering of the upper fall, with a good simulation of the DM content at PC (0.12 mg/L) by taking $d_3 = 0.49 \text{ day}^{-1}$.

Dissolved oxygen budget

Contrary to the DM, in direct link with the persistence of deoxygenated waters in the bottom layers of the reservoir, there has not been much improvement in the DO content of the turbined water which was equal to 1.3 mg/L on average in 2003, in the range of the mean yearly values of the 1995-2002 period (0.5-1.5 mg/L). It was shown that close to 90% of the upstream deficit to the DO saturation value (7.9 mg/L) was filled in by the aerating weir- in its initial configuration - for the usual range of turbined discharges, generating DO concentrations greater than 7 mg/L at the outlet of the weir. Under such conditions, 0.8 km downstream of the dam, the monthly DO concentrations measured are slightly lower, with mean annual values in the range 6.8-7.0 mg/L for each year of the 1995-2001 period, and logically, with a mean value of 6.0 mg/L and 5.0 mg/L in 2002 and 2003 respectively, after the 2 successive reductions of the upper fall of the weir which have reduced the atmospheric inputs of oxygen. The lowest concentrations at this point are observed when the turbined discharges are close to 100 m3/s (giving more time for the expression of the DO sinks) or when the bottom gates are open simultaneously (reducing the global efficiency of aeration downstream of the dam). Due to the oxidation of DM and other compounds released by the dam, and the very low inflows of the downstream tributaries, the DO content is decreasing progressively downstream, with over the March 1995-1998 period, a minimum generally reached between RB and PC and generally maintained down to PC. For very low turbined discharges (100 m³ s⁻¹ or less), the minimum was observed as soon as CV (Gosse et al 2002). At PC, for the usual range of turbined discharges (150-270 m³ s⁻¹), the manual monthly survey reveals a mean annual value of 3.1 mg/L in 1995, 3.5 mg/L in 1996, 4.1 mg/L in 1998, 5.0 mg/L in 1999, 5.7 mg/L in 2000 and 2001, 4.7 mg/L in 2002 after the first reduction of the weir, and 4.1 mg/L in 2003 after the second reduction. In relation to the seasonal variation in quality of the flow released by the dam, the DO concentrations at PC are slightly higher during the rainy season. The continuous recordings of DO at this station do not show a significant daily cycle, the average difference between the minimum and maximum being close to 0.5 mg/L. The analysis of the recording on the October 1997 - January 1998 indicates that this variation is mainly due to short peaks appearing regularly in the afternoon, which can reveal an influence of wind action or photosynthesis.

A question raised is the determination of the main factors explaining the change in the mean annual decrease of DO between the point situated 0.8km downstream of the dam and PC, decrease which was around 3.5 mg/L for years 1995-1998, close to 2 mg/L in 1999 and close to 1.2 mg/L for years 2000-2003. To answer, we can use the simplified DO/DM model indicated above which was developed for analysing the 1994-1998 data set. Due to a relatively low influence of photosynthesis over the period, the equation for DO was written (Gosse et al 2002): $dC / dt = K'_2 (Cs - C) - (D_1 + D_2) - f. k_3.DM_0 - BEN / H,$ with K'_2 aeration coefficient for DO (noted C in the equation) which is 1.1 greater than K_2 considering the ratio of the Schmidt numbers and the comparative efficiency of DM elimination and DO input at the weir, and f (gO2/g CH4) a stoechiometric coefficient giving the quantity of oxygen

consumed by unit of DM used by bacteria. D1 (mg 0_2 /L/day) is the rapid chemical consumption in a few hours of the reduced matter coming from the bottom of the reservoir (previously taken at 0.5 mg/l in 4 hours), D2 is the 2- day slower DO demand linked to the decay of the oxidizible matter coming from the reservoir (DM excluded) previously taken at 0.2 mg/L/day for 1998. BEN/H – assumed to be a benthic demand - was introduced in the model as the measured DO sinks not linked to DM oxidation - in particular the 2 day BOD measured downstream of the weir and at PC - could not systematically explain the total longitudinal decreases of DO . The model has 2 calibration parameters, BEN (g/m2/day) and f which is equal to 4 if it is considered that 2 moles of O2 are necessary for the degradation of one mole of CH4.

The challenge is the capacity of this model to reproduce the radically new situation of years 2000-2003 a period characterised by a much lower longitudinal decrease of DO. A first serious indication of possible success is given by the analysis of the DM budget (see above) which shows firstly that the velocity of methane oxidation has decreased significantly after 1999 and that consequently the higher content of DM in the lower part of the river is more subject to emission into the atmosphere due to higher winds in that zone. Another indication is the results of the 2-day BOD monthly measurements 0.8 km downstream of the dam and at PC, even if the extrapolation of such in vitro measurements must be prudent: at PC, the BOD values are 2 times greater on the 2000-2003 period (1.0 mg/L) by comparison to the 1995-1998 period during which there was almost no DM at that station (0.6 mg/), whereas they have decreased by 40% at the upstream point (from 3.0 mg/L in 1995-1998 to 1.8 mg/L in 2000-2003) with a probable influence of the reduction in the velocity of methane oxidation. If we run the model with the new formation of K'2 derived from the new formulation of K2 proposed above for DM, and keep the D₁ and D₂ values previously considered for 1998, we find that the choice f=4 and BEN=0.5 g/m2/day reproduces correctly at 220 m3/s the decrease from 5.0 mg/L downstream of the weir to 4.0 mg/L at PC observed on average (statistics on 7 profiles) in 2003 after the 2nd lowering of the weir in the usual range of daily turbined discharges. With this calibration, the model indicates that DM oxidation represents 68% of the DO sink (2.7 mg/L) taking place along the stretch and that the input of DO by aeration is 1.7 mg/L along the stretch. If we take f=3 which is biologically credible and generally fits better with in vitro BOD tests (Gosse et al 2002), BEN must be taken at 2.1 g/m2/day. That does not change the values of the total DO sink and the input of DO by aeration, but the proportion of methane oxidation in the DO sink which becomes equal at 51%. The choice of the 2 couples f/BEN gives also a correct DO concentration at PC (5.7 mg/L) for the turbined discharges of year 1999, with a proportion of DM oxidation in the DO sink equal to 59% or 43% depending on the f/BEN couple chosen, which are percentages logically lower than in 2003, as there were less DM in the water. The 2 coupes represents also rather well the decrease from to 6 mg/L (and 5.4 mg/L) at 0.8 km downstream of the dam to 4.5 mg/L (and 3.5 mg/L) at PC observed during the May (and December) 2003 GHG campaigns.

Dissolved carbon dioxide budget

Routine (monthly) and coherent data of DC are only available for the period 1999-2001. In the usual range of daily turbined discharges - 150-270 m³ s⁻¹ - they indicate - for 1999, 2000, 2001 respectively - a mean concentration of 27.5, 17.5 and 19.5 mg/L in the turbines, 18.5, 12 and 12.5 mg/L 0.7 km downstream of the aerating weir, and 15.5, 11 and 10.5 mg/L at PC. These results show the aeration effect of the weir and reveal a decrease of around 2 mg/L of DC along the river. Which indicates that the internal production of CO2 by the different DO sinks is not sufficient to counterbalance the aeration processes at the air-water interface. It is thus very interesting to notice that - for these 3 years at least - almost identical mechanisms producing CO2 and consuming O2 increase the gap between the concentration of a dissolved gas to its saturation level in one case (O2) and reduce this gap for the other (CO2). A first explanation is that this gap is less than 5 mg/L in one case (O2) and more than 10 mg/L in the other (CO2), the higher the gap the higher the aeration effect plays. The GHG December campaign is of interest to give an insight on the order of magnitude of the DC budget in the river, as there is for this campaign a rather acceptable coherence between the aeration coefficients calculated independently from CO2 and CH4 data (there is presently a problem of coherence to be resolved for CO2 for the campaign of May 2003 between the emission flux to the atmosphere and the concentrations measured in the river as they give for at least 2 points an aeration coefficient 3 times greater than expected by comparison to the CH4 aeration coefficient). It appears that the December campaign suggests a decrease of 1-2 mg/L of DC (from 15-16 mg/L to 14 mg/L) from the outlet of the weir to PC for an emission of CO2 to the atmosphere measured at 40 g/m2/day on average along the stretch.

Let us apply the simplified modified indicated earlier by adding a 3rd equation for simulating DC (written X here in mg/L). The equation can be written: dX / dt = K"₂ (Xs - X) + 1.375 ($(D_1 + D_2) + f. k_3.DM_0 + BEN / H$). It does not add any calibration parameter. K"₂, the aeration coefficient for DC is - as indicated earlier - deduced as indicated earlier from K_2 using the ratio of Schmidt numbers, and it is considered that a mole of CO2 is produced when a mole of O2 is consumed inside the river. Xs is taken equal to 0.5 mg/L which characterizes most of the stretch. If we do the same simulation than for CH4 and DO consisting at describing the mean situation of the usual range of turbined discharges – with 220 m3/s – in 2003 after the 2^{nd} lowering of the weir, we have to specify the DC at the outlet of the weir : we have at our disposal only 2 profiles - in August and September 2003 – which give pertinent results from upstream the weir down to PC with a mean value of 19 mg/L at the outlet of the weir and 17 mg/L at PC. Theses

values appear close to those given by the profile of the GHG campaign of December 2003 which suggests a decrease of 1-2 mg/L of DC (from 15-16 mg/L to 14 mg/L), although the turbined discharge was only 115 m3/s during the campaign. If we run the model with a value of 19 mg/L for DC at the outlet of the weir, the model gives a concentration of DC at PC equal to 15 mg/L, which is slightly lower than expected according to the 2003 data but rather in accordance with the richer data set of 1999 for an equivalent mean level of DC downstream of the weir. The model gives CO2 emissions to the atmosphere from 22 g/m2/day in the upstream part of the stretch to 38 g/m2/day in the downstream part, levels which are in phase with those measured by the Québec team in 2003. The model indicates that under such conditions, the internal production of CO2 from oxidation processes along the 40 km stretch (12 g/m2/day on average) replaces close to half of the flux of CO2 emitted by aeration from the stretch to the atmosphere.

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