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Different regulation of CO_2 emission from streams and lakes

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Abstract

It has become more and more evident that CO_2 emission (F_{CO_2}) from freshwater systems is an important part in the global carbon cycle. Only few studies addressed the different mechanisms regulating F_{CO_2} from lotic and lentic systems. In a comparative study we investigated how different biogeochemical and physical factors can affect F_{CO_2} from streams and reservoirs. We examined the seasonal variability in CO_2 concentrations and emissions from four streams and two pre-dams of a large drinking water reservoir located in the same catchment, and compared them with parallel measured environmental factors. All streams generally were supersaturated with CO_2 over the whole year, while both reservoirs where CO_2 sinks during summer stratification and sources after circulation. F_{CO_2} from streams ranged from 23 to 355 mmol m^{-2} d^{-1} and exceeded the fluxes from the reservoirs (−24 to 97 mmol m^{-2} d^{-1}). Both the generally high piston velocity (k) and CO_2 oversaturation were responsible for the higher F_{CO_2} from streams in comparison to lakes. In both, streams and reservoirs F_{CO_2} was mainly controlled by the CO_2 concentration (r = 0.86 for dams, r = 0.90 for streams), which was clearly affected by metabolism and nutrients in both systems. Besides CO_2 concentration, also physical factors control F_{CO_2} in lakes and streams. During stratification F_{CO_2} in both pre-dams was controlled by primary production in the epilimnion, which led to a decrease of F_{CO_2}. During circulation when CO_2 from the hypolimnion was mixed with the epilimnion and the organic matter mineralisation was more relevant, F_{CO_2} increased. F_{CO_2} from streams was physically controlled especially by geomorphological and hydrological factors regulating k, which is less relevant in low wind lakes. We developed a schematic model describing the role of the different regulation mechanism on F_{CO_2} from streams and lakes.

Taken together, F_{CO_2} is generally mostly controlled by CO_2 concentration in the surface water. Lake stratification is a very important factor regulating F_{CO_2} from lakes via controlling CO_2 concentration and metabolism. But F_{CO_2} in heterotrophic streams is generally higher. The higher k values are responsible for the comparable high F_{CO_2}.

BGD
10, 10021–10053, 2013

On a Central European landscape scale CO_2 emission from streams was more relevant than the CO_2 flux from standing waters.

1 Introduction

Gaseous CO_2 emission from inland waters is an important component of the global carbon cycle (Cole et al., 2007). The CO_2 flux between water and atmosphere is by diffusion across the air–water interface which depends on the CO_2 concentration difference between water and atmosphere and a transport coefficient (piston velocity, k). Typically surface waters are oversaturated with respect to CO_2, making them a CO_2 source. For European lakes, a mean CO_2 emission of 24 $\mathrm{mmol\,m^{-2}\,d^{-1}}$ was estimated which could give a total emission of 17 $\mathrm{Mio\,t\,yr^{-1}}$ from all European lakes (Kastowski, 2011). Lots of data are available from lakes and reservoirs (reviewed e.g. in Tremblay et al., 2005; Barros et al., 2011) or from rivers and streams (Wanninkhof et al., 1990; Owens et al., 1964), but only few studies combine both lake and river systems (Guerin et al., 2007; Jonsson et al., 2007).

The greenhouse gas (GHG) concentration and emission from freshwater systems is controlled by different environmental factors and by internal processes. Thereby seems the majority of the CO_2 in lakes or streams to originate from organic terrestrial sources (Sobek et al., 2003; Humborg et al., 2010). The mineralisation of terrestrially originated dissolved organic carbon (DOC) is often considered as the main source for a CO_2 oversaturation, mostly found in boreal lakes (Sobek et al., 2003). Although DOC seems to be a predictor of pCO_2 in many lakes, the shape of the relationship varies greatly among regions (Roehm et al., 2009). In a Finnish lake study, where pCO_2 was elevated in agricultural catchments, it was strongly associated to total nitrogen (TN) and total phosphorus (TP) but not with total organic carbon (TOC) (Rantakari and Kortelainen, 2008; Kortelainen et al., 2006). In a long term study in 37 large Finnish lakes CO_2 emission (F_{CO_2}) was closely related to the annual precipitation pattern (Rantakari and

BGD
10, 10021–10053, 2013

Kortelainen, 2005) while there were only weak correlations to water chemistry, TOC or land use in the catchment.

CO_2 evasion could also depend on lake area. In very small and very large lakes negative relations with lake size coupled to several characteristics (depth, land use characteristic, etc.) were found (Kelly et al., 2001). Especially in small shallow lakes sediment respiration affects CO_2 concentration (Kortelainen et al., 2006). Metabolic processes can generally affect CO_2 concentration in lakes. Primary production consumes CO_2 and thus, there are several studies showing that a higher trophic state reduces CO_2 emission (Trolle et al., 2012). Nutrient rich eutrophic lakes may even be undersaturated with CO_2 making them a CO_2 sink rather than a source (Balmer and Downing, 2011). However, the seasonal variability of CO_2 concentrations is highly synchronous to lake stratification. Accumulation of CO_2 in the hypolimnion during stratification leads to an increase of CO_2 concentration in the upper water during lake mixing (Kortelainen et al., 2006).

Factors regulating CO_2 emission from streams could be the same as those influencing the emission from lakes. Recent studies showed that GHG emission from streams or rivers could – likewise to lakes – be affected by pH, temperature, several nutrients, CO_2 concentration itself and general hydrological or geomorphological conditions (Alin et al., 2011; Wallin et al., 2011; Rantakari, 2010; Li et al., 2012).

In the Yangtze River CO_2 outgassing was controlled by the pH of the water (Li et al., 2012). The pH controls mainly the speciation of dissolved inorganic carbon (DIC) and therefore also the CO_2 concentration which should be directly affect its emission (Neal et al., 1998). Genereux and Hemond (1992) described that CO_2 degasing is linked to velocity and turbulences. Thus especially small turbulent streams tend to emit large amounts of CO_2. Of course, also groundwater DIC input regulates CO_2 concentration, especially in small streams, and affects therefore also emission (Battin et al., 2008). Different adjacent soil or sediment types might have different water storage periods regulating the DIC accumulation time (Rantakari, 2010).

BGD
10, 10021–10053, 2013

Even if controlling factors seem to be often similar in both systems, several studies showed for streams higher CO_2 fluxes than for lakes. Teodoru et al. (2009) estimated for streams located in the north western boreal region a daily CO_2 emission between 58 and 250 mmol C m^{-2}. This could be up to two fold higher than emissions from lakes in the same region. We assume that the different regulation mechanisms are different relevant for lakes or streams. This could be an important issue if GHG emission on catchment scale is studied, because one would expect streams and lakes to react differently to climate and/or landuse change. It becomes clear, that quantification of GHG emission from freshwater systems on a catchment scale must include lotic and lentic systems, as well as the different land use form of the investigated region.

To our knowledge no studies exist where the factors influencing GHG emission in lakes and streams are directly compared in a temperate ecosystem. The number of studies where CO_2 evasion from both streams and lakes located in one catchment was investigated is rather rare or only available from boreal catchments. By measuring the CO_2 flux from 4 streams and 2 reservoirs in the same catchment in a typical central European setting we wanted to find out, whether streams or lakes emit more CO_2 per area and what are the underlying reasons for that. We hypothesize that in the temperate zone both systems are affected by the same environmental factors, but with different intensities. By analysing seasonal trends and correlations with various environmental parameters, we want to identify and compare the mechanisms controlling the CO_2 flux from lotic and lentic waters.

2 Materials and methods

2.1 Study sites

The study sites are located in the upper part of the Bode catchment in the Harz Mountains, Central Germany (Fig. 1). Two of the investigated streams are pristine streams (Ochsenbach and Zillierbach) located next to the Harz National Park while two other

BGD
10, 10021–10053, 2013

streams were running through more rural areas (Hassel and Rappbode). A detailed description of these streams is given in Halbedel et al. (2012). They are parts of a stream network that drains into the Rappbode reservoir system (Rinke et al., 2013). The Hassel as well as the Rappbode is draining directly into the respective pre-dams Hassel (DH) and Rappbode (DR), which were also investigated in this study. DR is a mesotrophic, DH a eutrophic softwater reservoir. Since their water level is not regulated and the outflow is allways over the dam, pre-dams are suitable model systems for lakes. The general characteristics of the investigated sites are given in Table 1. Detailed information about the chemical characteristic of streams located in the Bode catchment and about the prevalent land use forms are presented in Kamjunke et al. (2013).

2.2 Field work

2.2.1 Pre-dams

Both pre-dams were sampled biweekly to monthly at a routine monitoring site at the deepest point close to the dam. Samples for routine water analysis of the pre-dam water were taken using a Ruttner water sampler (Limnos, Finland). For CO_2 analysis, glass vials were half filled and closed with a rubber septum. To correct for ambient CO_2 in the headspace, ambient air samples were taken in separate vials. From November 2011 samples were taken with 60 mL syringes closed by a 3-way stop cock. Ambient air samples were also taken with the same type of syringes. Syringes were only filled half, stored cool and analyzed within 24 h in the laboratory. Prior to analysis, a gas headspace of 30 mL N_2 was added to the syringes and the syringes were shaken on a rotary shaker for 30 min. Vertical profiles of temperature (T), O_2, and pH were measured with a multiparamter probe (Ocean-Seven, Idronaut, Italy).

BGD
10, 10021–10053, 2013

2.2.2 Streams

The streams were sampled in spring, summer and autumn 2011. Each CO_2 sampling campaign was at base flow for one to two days. The CO_2 measurements were done simultaneously to whole stream metabolism measurements, which are described in Halbedel et al. (2012). Detailed information about the collection of several environmental parameters like reaeration coefficient ($k_{propane}$), discharge (Q), lateral inflow (l), width (w) and depth (d) of the stream reach, velocity (v), reach length, travel time (t), pH, conductivity (cond.), and oxygen (O_2) can also be found there. There is also described how samples for further chemical analysis were collected (ammonium (NH_4^+), nitrate (NO_3^-), total phosphorus (TP)). All chemical samples were taken twice a day, at noon and one hour before sunrise. We expected highest primary production at noon and no primary production before sunrise. Water samples for chemical analysis were taken with the "wave" in the thalweg, at the in- and outflow of the stream reach. For chlorophyll *a* (Chl *a*) analysis water was filtered (GF/F, 45 µm pore size) directly in the field and filter were immediately frozen in liquid nitrogen and stored at −20 °C. For TIC water samples were collected directly below the water surface without air bubbles. The local atmospheric pressure (p, mmHg) and the stream near atmospheric temperature (T_{air}, °C) were measured with a handheld barometer or thermometer, respectively.

For CO_2 measurements, water samples were taken at a defined stream reach (cp. Halbedel et al., 2012) following the "wave". The reach in- and outflow and seven additional positions that were consistently distributed over the investigated stream reach were sampled. The water was collected by the use of a 60 mL plastic syringe from a depth of approximately 10 cm below the stream surface in the thalweg and equilibrated with headspace of ambient air by vigorous shaking for 1 min below the water surface (Kling, 1991; Hope et al., 2004). The equilibrated air was then injected in 12 mL evacuated crimp vials. Three additional samples of ambient air were taken at the in- and outflow and in the middle of the stream. All vials were stored at 4 °C until analysis, which were conducted within 48 h in the laboratory.

BGD
10, 10021–10053, 2013

2.3 Analytics

Equilibrated air of the stream samples, headspace gas from lake samples, and all environmental air samples were analyzed with a SRI 8610C gas chromatograph equipped with a flame ionization detector. From the gas concentration in equilibrated headspace samples, pressure and temperature the concentrations of CO_2 in the water (mmol L^{-1}) was calculated by applying Henry's law (Kling et al., 1991).

NO_3^- and NH_4^+ were determined photometrically applying the segmented flow technique (Halbedel et al., 2012). Total phospor (TP) was measured using the ammonium molybdate spectrometric method (Halbedel et al., 2012). Total inorganic carbon (TIC) and dissolved organic carbon (DOC) were analysed based on high temperature oxidation with NDIR-detection (Kamjunke et al., 2013). Chlorophyll *a* (Chl *a*) was measured by HPLC (DIONEX Corporation, Germany) using the ethanol extraction method (Koschorreck and Wendt-Potthoff, 2012).

2.4 Calculations

2.4.1 Flux calculations

The CO_2 flux between the water surface and the atmosphere (F_{CO_2}) was determined from the difference between the actual CO_2 concentration in the surface water (C_{2water}) and the concentration in air equilibrated water (C_0) multiplied by the gas transfer velocity (k):

$$F_{CO_2} = (CO_{2water} - C_0) \times k \quad (1)$$

C_0 was calculated from the CO_2 partial pressure in the ambient air samples using Henry's law.

The k was determined differently for reservoirs and streams. For reservoirs k (in this case k_{600}) was calculated from wind speed and normalized to a Schmidt number of

BGD
10, 10021–10053, 2013

600 (Crusius and Wanninkhof, 2003):

$$k_{600} = \left[1.68 + \left(0.228 \times U_{10}^{2.2}\right)\right] \times \left(\frac{SC_{CO_2}}{600}\right)^{-0.5} \quad (2)$$

U_{10} is the wind speed at 10 m above the surface ($m\,s^{-1}$) and was calculated from the mean wind measured by a hand held anemometer at 1 m ($U = 3 \pm 1.4\,m\,s^{-1}$, $n = 22$):

$$U_{10} = U \times 1.22 \quad (3)$$

The Schmidt number SC_{CO_2} was calculated from the surface water temperature T (°C):

$$SC_{CO_2} = 1911.1 + (118.11 \times T) + (3.4527 \times T^2) - (0.04132 \times T^3) \quad (4)$$

The k for streams was calculated from gas transfer coefficients for propane ($k_{propane}$) obtained from parallel metabolism studies (Halbedel et al., 2012), which were converted to k_{CO_2} (Genereux and Hemond, 1992):

$$k_{CO_2} = k_{propane} \times \left(\frac{d_{CO_2}}{d_{propane}}\right)^{n} \quad (5)$$

The exponent n can potentially vary from −0.66 and −0.5. We use −0.5 that was given in Hope et al. (2001). d_{CO_2} and $d_{propane}$ were calculated for the actual stream temperature (in °C) using the following equations (Hope et al., 2001):

$$d_{CO_2} = 1.005 \times \exp(0.00231 \times T) \quad \text{and} \quad d_{propane} = 1.092 \times \exp(0.0235 \times T) \quad (6)$$

k was than calculated with Eq. (7):

$$k = k_{CO_2} \times t \times \frac{Q}{A} \quad (7)$$

with t = travel time, A = stream reach surface (m^2), which was calculated from mean width and reach length (data from Halbedel et al., 2012).

BGD
10, 10021–10053, 2013

2.4.2 Statistics

The significance of correlations was tested with the Spearman rank order correlation. The non-parametric Wilcoxon test was used to test the significance of differences between groups. All statistical analyses were conducted in SigmaPlot 12.0.

3 Results

3.1 CO_2 concentrations and evasion

All investigated streams were supersaturated with CO_2. With values ranging from 28 to 200 µmol L^{-1}, the CO_2 concentrations in the investigated streams were mostly higher than in the reservoirs, which had values ranging from 0 to 131 µmol L^{-1} (Fig. 2). A wide scatter of data was found for the Hassel stream, which had significantly the highest CO_2 concentrations in general (median: 109 µmol L^{-1}). The data scatter for CO_2 was small in the other three streams. The CO_2 concentrations were similar in both pre-dams.

Values for k_{CO_2} in the streams ranged from 0.01 to 0.05 min^{-1}. The gas transfer coefficients were negatively correlated to Q ($r = -0.79$). Also the calculated k values were in all streams higher than in the reservoirs (Table 1). Both reservoirs had the same k value and the CO_2 flux from both reservoirs was also similar (Fig. 3). The F_{CO_2} from streams was between 23 to 355 mmol m^{-2} d^{-1}. These fluxes are higher than the evasion calculated for the pre-dams (from −24 to 97 mmol m^{-2} d^{-1}). Whilst the CO_2 evasion from both reservoirs was in the same range, the streams had more variable emission values. With a median of 251 mmol m^{-2} d^{-1} the Hassel had by far the highest CO_2 emission rate; while the other streams had lower (but still higher than the reservoirs) area specific emission rates (Fig. 3). We estimated the yearly CO_2 emission from the different waters based on means and surface area: 4.06×10^{-6} kmol yr^{-1} for Hassel, 1.73×10^{-6} kmol yr^{-1} for Rappbode, 5.61×10^{-4} kmol yr^{-1} for Ochsenbach, and 2.11×10^{-5} kmol yr^{-1} for Zillierbach; and for the reservoirs: 5.69×10^{-2} kmol yr^{-1} for

BGD
10, 10021–10053, 2013

DH and 1.58×10^{-3} kmol yr^{-1} for DR. We assume that CO_2 flux is negligible during the winter months when both waters can be covered by ice.

3.2 Seasonality

A more detailed picture is given if looking on the seasonal changes of the GHG data (Fig. 4a). The CO_2 concentrations were especially in Ochsenbach and Zillierbach, but also in the Rappbode nearly constant over the whole year. We calculated median values for each season. Medians for these streams ranged from 45 to 51 µmol L^{-1} for the Zillierbach, from 44 to 54 µmol L^{-1} for the Ochsenbach, and from 56 to 59 µmol L^{-1} for the Rappbode. Compared to all the other sites the Hassel had the highest CO_2 concentrations and fluxes (Fig. 4b) as well as the most pronounced differences between seasons. Highest concentrations of 176 µmol L^{-1} were observed in fall. In comparison, the Rappbode had with 126 mmol m^{-2} d^{-1} (median) the highest CO_2 evasion rate in spring. The evation decreased in summer and fall to 85 and 69 mmol m^{-2} d^{-1} (median values). This decrease in F_{CO_2} was also found for the two pristine streams Zillierbach and Ochsenbach. The median values ranged for the Zillierbach from 75 mmol m^{-2} d^{-1} in spring to 34 mmol m^{-2} d^{-1} in fall, and for the Ochsenbach from 85 mmol m^{-2} d^{-1} in spring to 51 mmol m^{-2} d^{-1} in fall.

Similar to the Hassel, both reservoirs exhibited a pronounced seasonality of the CO_2 concentration and CO_2 fluxes with low surface values during spring and summer, and high values during autumn when the data were also more variable (Fig. 4). The reservoirs were stratified from March until November. During the stratification period the surface water contained low concentrations of CO_2 while CO_2 accumulated in the bottom water to maximum concentrations of 400 µmol L^{-1} (Fig. 5). This is in the range of stream concentrations. Sometimes, during summer the reservoirs were even under saturated in the surface water. In fall, the bottom water was mixed into the epilimnion, leading to high surface concentrations and evasion rates.

BGD
10, 10021–10053, 2013

3.3 The role of environmental factors

Table 2 gives an overview of the correlations between different environmental parameters and CO_2 flux. Not surprising, CO_2 emission correlated significantly positively with CO_2 itself in streams and pre-dams. The CO_2 flux from these streams to the atmosphere can be predicted from the following regression: CO_2 flux = $-13.217 + (1.920 \times CO_2)$, with $p < 0.001$, and CO_2 emission from the reservoirs can be predicted from: CO_2 flux = $-16.139 + (0.666 \times CO_2)$, with $p < 0.001$. CO_2 evasion was also negatively correlated with temperature and pH in dams.

In the reservoirs, CO_2 emission was also correlated to Chl *a* and there was a significant correlation between Chl *a* and CO_2 concentration. The Chl *a* concentration in the reservoirs followed a typically seasonal trend with increasing concentrations from spring to summer and a decrease in autumn. The seasonal concentrations of parameters (Chl *a*, DOC, TIC, NH_4^+, NO_3^-, TP) that are associated with primary production or respiration are given in Fig. 6. In comparison to both pre-dams all investigated streams were less productive. This is indicated by low Chl *a* concentrations (Fig. 6c) and a general low gross primary production (GPP, data from Halbedel et al., 2012). There was a positive correlation between F_{CO_2} and total phosphorus (TP) detected for streams but not for reservoirs. TP was highest in the Hassel (Fig. 6f) and lowest in both forest streams, but also in the Rappbode reservoir. Slightly higher but still low concentrations were measured in Rappbode and in the Hassel reservoir. CO_2 emission from both systems correlated with ammonium concentration. Both forest streams and the Rappbode had low ammonium concentrations (Fig. 6d). The highest ammonium concentrations were measured in Hassel, in summer, after extensive cow pasture. In this stream ammonium was also slightly elevated in spring and fall. Both reservoirs had comparable high ammonium concentrations in autumn. During other periods the median of the ammonium concentrations of the pre-dams were in the range of the other stream values. The CO_2 flux from streams correlated also positively with TIC. Thereby had Hassel the highest TIC concentrations. The lowest values were measured there in summer and

BGD
10, 10021–10053, 2013

the highest in spring and fall (Fig. 6b). Also high TIC concentrations were detected for Rappbode, where highest values were found in summer and lowest values were found in fall. Zillierbach showed highest TIC concentrations in summer, but lowest were detected in spring. The lowest TIC was measured for Ochsenbach. The TIC increased in both pre-dams continuously from spring to autumn. We found no other significant correlations between CO_2 evasion and environmental factors, especially not with DOC or NO_3^-.

4 Discussion

The CO_2 evasion per m^2 from streams exceeded the CO_2 emission from the reservoirs by more than one order of magnitude (Fig. 3). As was recently shown by Knoll et al. (2013), especially older reservoirs could temporary even be CO_2 sinks in a temperate landscape. On the other hand, streams are generally known to be CO_2 sources rather than sinks (Teodoru et al., 2009; Wallin et al., 2012). Why do CO_2 emissions from streams and rivers outreach CO_2 flux from lakes per area? As shown in Eq. (1), CO_2 emissions from streams and lakes depend both on the surface concentration of CO_2, which is probably primarily regulated by biogeochemical processes, and the physical transfer coefficient k. It is the question, whether both factors are equally important in the two types of aquatic systems.

The mean emission from the reservoirs was $26\,g\,C\,m^{-2}\,yr^{-1}$ for DH and $79\,g\,C\,m^{-2}\,yr^{-1}$ for DR. On an annual basis, both reservoirs were small CO_2 sources. The reservoirs, however, were seasonally undersaturated and therefore temporary CO_2 sinks. The mean emission from the eutrophic DH was in the range presented by Knoll (2013) for two reservoirs located in the temperate zone in the USA (11.5–$33.6\,g\,C\,m^{-2}\,yr^{-1}$). The higher annual CO_2 emission in DR could be related to the lower nutrient loading of this mesotrophic reservoir resulting in lower primary production.

In contrast to the reservoirs, all investigated streams were supersaturated with CO_2. The CO_2 oversaturation measured in all streams indicates the general heterotrophic

BGD
10, 10021–10053, 2013

stream character. We recently showed that all these streams were net heterotrophic (Halbedel et al., 2012). A recent study in the same catchment found all investigated streams and rivers in this landscape also supersaturated (Kamjunke et al., 2013). Most streams draining temperate or boreal regions, arctic tundra, peatlands and tropical ecosystems are supersaturated with CO_2 (Richey et al., 2002; Hope et al., 2001, 2004; Rantakari, 2010; Butman and Raymond, 2011; Wallin et al., 2010).

The mean CO_2 concentration in our streams was 70 µmol L^{-1} compared to 37 µmol L^{-1} in the reservoirs. Why did the streams have higher CO_2 concentrations than the reservoirs? The CO_2 oversaturation in the streams requires a strong CO_2 source, considering that equilibration with the atmosphere should be faster in moving waters. Especially in streams, groundwater inflow might be a significant CO_2 source (Humborg et al., 2010). The highest lateral inflow (> 15 % of Q) was detected for the stream Hassel (data from Halbedel et al., 2012). This stream drains a peatland, suggesting CO_2 concentration and emission is directly affected by the adjacent peatland. The groundwater inflow to the other streams was much lower and sometimes rather an outflow than an inflow. We think that these streams were not significantly affected by groundwater. Even though we have not investigated the groundwater inflow into to the reservoirs we think, because of their geological underground (bedrock) it is not directly affecting the CO_2 evasion. We conclude that groundwater had a minor influence on CO_2 emissions in our study. In aquatic ecosystems CO_2 derives from the mineralisation of organic matter. As shown below, the physical separation of the zone of organic matter mineralisation from the water surface is probably a major reason for the lower surface CO_2 in the reservoirs.

The comparison of gas transfer velocities in lentic and lotic waters is hampered due to the different methods used in river and lake research. Traditionally, in lake research the transfer velocity is expressed as k (or k_{600}) having the unit of a velocity (m s^{-1}). The analogue parameter in river research is called "reaeration coefficient" (k_{CO_2}), having the unit m^{-1}. Both parameters are related by Eq. (7). Thus, the conversion of k_{CO_2} to k requires data on travel time, discharge and stream area. Especially, the precise

BGD
10, 10021–10053, 2013

measurement of stream area is not trivial and introduces an additional source of error in the determination of k in streams. We also tested the approach of Alin et al. (2011), which is based on depth values, which lead to a significant increase of most of the values, indicating the high sensitivity towards the hydromorphological data. However, the comparison of literature values is difficult, especially if no travel time, stream area, depth or discharge data are provided.

In standing waters, k depends on surface turbulence which in turn depends on the weather conditions (wind, precipitation). In the reservoirs we assumed that k is proportional to wind speed and used a fixed mean wind speed of $3\,m\,s^{-1}$ for our calculations. At such low wind speed k is usually rather constant and not depending on wind. Typically, below a wind speed of $5\,m\,s^{-1}$, k values fluctuate in a narrow range between 1 and $6\,cm\,h^{-1}$ (Crusius and Wanninkhof, 2003). Only short periods of higher wind intensities (Crusius and Wanninkhof, 2003) or precipitation (Cole and Caraco, 1998) may lead to episodic higher k, but the resolution of our measurements was not high enough to resolve the effect of local short term wind fluctuations on CO_2 emission. Since under our low wind setting, k is probably not directly related to wind speed, and we did not have continuous on-site wind data, we decided to use a constant k to calculate CO_2 fluxes from the reservoirs.

In the streams k was on average twofold higher than in the reservoirs. The k values were in the upper range of those published in Allin et al. (2011), which are based on different k_{CO_2} values from literature, as well as their own data. A reason for our comparable low values could be that in their study k was calculated based on depth values. The k_{CO_2} values detected for the Harz Mountain streams are in the range of those published for boreal headwater streams ($0.001–0.207\,m^{-1}$, Wallin et al., 2011) and temperate peatland streams ($0.015–0.344\,m^{-1}$, Hope et al., 2001).

Thus, it turned out that both the CO_2 concentration and the transfer coefficient were higher in streams than in the reservoirs. Since both the mean CO_2 concentration and k were twofold higher in the streams, we conclude that both factors are equally responsible for the higher areal CO_2 emission from streams.

BGD
10, 10021–10053, 2013

Knowing the reasons for the absolute higher CO_2 emission fluxes from streams, the question is how the CO_2 flux is regulated in the two systems. We may analyze the seasonal dynamics as well as the correlation with different environmental parameters. The observed correlations between CO_2 flux and surface concentration suggest that the CO_2 concentration is the dominant factor in both systems. In case of the reservoirs this is not surprising since we used a constant k in our low wind setting. The CO_2 concentration, on the other hand showed a high variability in both reservoirs (39 ± 40 and $44 \pm 36\,\mu mol\,L^{-1}$ in the Hassel and Rappbode reservoir, respectively), resulting in a high variability of F_{CO_2}. The CO_2 concentration in the surface water is a result of the balance between CO_2 consuming primary production, the respiratory mineralization of organic matter and the physical gas transport. At least during summer, the surface CO_2 concentration and therefore also its flux from the reservoirs was controlled by primary production. This is supported by the correlation of the CO_2 flux with pH and Chl *a*. Many authors suggested that natural lentic systems with high primary production are sinks for CO_2 (Cole et al., 2007; Downing et al., 2008; Tranvik et al., 2009). In contrast, Knoll et al. (2013) showed recently that also productive reservoirs could be small CO_2 sources on a landscape scale. They found that reservoirs could be sinks only during dry summers and concluded a weather related summer difference in their net autotrophic lakes which is in accordance with previous findings (Cole and Caraco, 1998; Rantakari and Kortelainen, 2005).

In the streams, there was no correlation between Chl *a* or GPP and F_{CO_2} indicating that respiration is more relevant for F_{CO_2} from streams than primary production. The high impact of respiration on CO_2 emission is also shown by the correlation of F_{CO_2} with ammonium and phosphorus, which are products of the mineralization of organic matter (cp. Tranvik and Kokalj, 1998). The different chemical nitrogen forms as well as phosphorus were already shown to correlate with CO_2 evasion or CO_2 concentration in streams (Teodoru et al., 2009; Neal et al., 1998) and lakes (Kortelainen et al., 2000). Thus metabolism is generally a controller of CO_2 concentration and flux in both water systems, whilst nutrients could be indicators but also controllers of metabolism.

BGD
10, 10021–10053, 2013

There were no correlations between CO_2 evasion and DOC, neither in streams nor in lakes. This deviates from results of several studies on boreal lakes and streams where especially the turnover of organic carbon with terrestrial origin is considered as the main source for the CO_2 oversaturation (Sobek et al., 2003; Prairie et al., 2002; Jonsson et al., 2003; Dawson et al., 2009; and many more). However, particulate organic matter (POM) like seston, soil, sediment, litter and wood could also fuel the heterotrophic activity in waters (Rugenski et al., 2012; Vannote et al., 1980). Although POM variability wasn't investigated in this study, we assume that especially the heterotrophic turnover in both water systems is affected by POM.

However, besides metabolism also physical processes could have a significant impact on the CO_2 concentration. In stratified lakes, the zone of CO_2 consumption (epilimnion) is physically separated from the zone of CO_2 production (hypolimnion) (Boehrer and Schultze, 2008). This results in a depletion of CO_2 at the surface and an accumulation of CO_2 at depth. In the streams, these two zones do not exist and pelagial and benthal are closely coupled during the whole year. Thus, the standing waters can be temporary CO_2 sinks although being net heterotrophic while in streams net heterotrophy is always indicated by CO_2 oversaturation. As a result, lakes typically show highest CO_2 emissions during overturn when CO_2 rich bottom water is mixed to the surface (Kortelainen et al., 2000). Thus, the seasonal patterns of F_{CO_2} in reservoirs are controlled by physical processes rather than the rates of biogeochemical reactions. In contrast to this are the seasonal patterns in streams rather controlled by biogeochemical factors.

When k is rather constant or weather (wind, precipitation) controlled in lentic waters, then it is controlled by hydrodynamic factors in streams and rivers (Alin et al., 2011). We found that k_{CO_2} was negatively correlated to discharge indicating a general decrease of k_{CO_2} with increasing Q. There exist different findings in the literature regarding the linkage between k_{CO_2} and Q. Wallin et al. (2011) found for example no clear evidences for a coupling of k_{CO_2} and Q. They concluded that the impact on the variability of k_{CO_2} is highly site specific. Discharge is generally under suspicion to control the CO_2 flux from

BGD
10, 10021–10053, 2013

stream water to the atmosphere (Hope et al., 2001; Roberts et al., 2007). But we found no correlation between Q and F_{CO_2}, even though the higher ordering streams emit the highest CO_2 amount. Although slope was not measured in our study site, it has to be assumed that slope changes could generally affect the gas transfer. Wallin et al. (2011) indicated for boreal streams a general impact of slope on k_{CO_2}, which is supported by the results of earlier studies (Bennett and Rathbun, 1972; Gualtieri et al., 2002). There exist also studies showing that the geometry factors width and depth could correlate with k_{CO_2} (Wanninkhof et al., 1990; Genereux and Hemond, 1992). Thus, in streams k seems to be controlled by hydrodynamic factors, which can be very site specific.

It becomes clear, that the interplay of F_{CO_2} regulation factors is very complex. We developed a schema summing all relevant mechanisms (concentration, metabolism, lake stratification, k) together and explaining their effect on F_{CO_2} (Fig. 7). However, CO_2 concentration seems to be the most important factor regulating CO_2 flux from both lentic and lotic systems. Taking into account that CO_2 emission from streams is directly linked to the CO_2 concentration we estimated from the calculated linear CO_2 concentration-evasion relationship and the mean CO_2 concentration of the whole Bode catchment (data from Kamjunke et al., 2013) an annual CO_2 emission from the streams of 38.3 mol m^{-2} yr^{-1} for the whole Bode catchment (3229 km^2, 169 km length). Under the assumption of a mean stream width of 4 m a CO_2 flux from the water to the atmosphere of 2.59×10^4 kmol yr^{-1} can be estimated. This exceeds the CO_2 emission that can be assumed from lakes in the whole Bode catchment by far. Although a complete GHG budget for the catchment requires more detailed studies (e.g. outgassing at the dam, stream emissions during flood events), these estimation clearly shows the dominance of lotic systems for CO_2 emission on a Central European landscape level. Furthermore, the complexity of factors controlling CO_2 concentration and k in lakes and streams make it difficult to estimate future effects of land-use changes and climate changes on CO_2 emission from both types of waters. On may speculate that CO_2 emission from lakes will be more affected by climate change, while CO_2 emission from streams should be more affected by land use change and matter import from the

BGD
10, 10021–10053, 2013

catchment. Our results from the Hassel stream suggest that higher nutrient input from agricultural catchments lead to higher and more variable CO_2 emissions from streams. More comparative studies in different climate zones and landscapes could contribute to the understanding of these different systems.

5 Conclusions

The variability of CO_2 emissions from streams and reservoirs seems to be mainly controlled by the CO_2 concentration which is variable in time and affected by different environmental factors. Metabolism, which is known to be affected by nutrient availability and weather conditions, seems to control the CO_2 concentration and flux to the atmosphere in reservoirs and in streams. Whilst standing waters are stratified, primary production and organic matter mineralization are uncoupled, which leads to a strong control of primary production on the CO_2 flux from the water surface to the atmosphere. During circulation heterotrophic turnover controls CO_2 concentration and F_{CO_2} increase. Heterotrophic streams are controlled over the whole year by respiratory processes and thus, have a higher CO_2 concentration. Even if CO_2 concentrations in streams and lentic waters are in the same range during circulation, F_{CO_2} is in streams generally higher. The higher k values are responsible for the comparable high CO_2 emission. Thus, the annual CO_2 emission from temperate streams exceeds by far the evasion from temperate lentic waters. Although on a first glance the stream area is small, their CO_2 emission can affect the regional C balance on a landscape level.

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BGD
10, 10021–10053, 2013

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BGD
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BGD
10, 10021–10053, 2013

Table 1. Characterization of the investigated streams and reservoirs*.

	depth m	area km^2	volume Mio m^3	Q $L\,s^{-1}$	v $m\,s^{-1}$	pH	k $cm\,h^{-1}$
PD Rappbode	5.73	0.24	1.66	n.d.	n.d.	7.76	5.6
PD Hassel	5.03	0.26	1.64	n.d.	n.d.	8.33	5.6
Rappbode	0.26	46.52	n.d.	30.38	0.059	7.80	9.5
Hassel	0.10	43.13	n.d.	3.23	0.029	7.66	19.8
Zillierbach	0.10	10.70	n.d.	2.38	0.021	7.66	18.7
Ochsenbach	0.09	2.26	n.d.	3.45	0.034	7.31	14.8

* Data are means. Q = discharge, v = velocity, k = CO_2 gas transfer velocity, n.d. = not determined.

BGD
10, 10021–10053, 2013

Table 2. Correlation of CO_2 evasion with different parameters, sorted for dams and streams*.

	parameter	*r*	*p*	*n*
dams	**CO_2**	**0.86**	**< 0.001**	**34**
	Temperature	**−0.52**	**0.002**	**34**
	O_2	0.21	0.228	34
	pH	**−0.75**	**0.000**	**34**
	Chl *a*	**−0.45**	**0.011**	**31**
	DOC	0.09	0.620	34
	TIC	0.13	0.448	34
	NH_4^+	**0.47**	**0.006**	**33**
	NO_3^-	−0.09	0.611	31
	TP	−0.25	0.157	34
	cond.	−0.24	0.163	34
streams	**CO_2**	**0.90**	**< 0.001**	**209**
	Temperature	0.41	0.173	12
	O_2	−0.25	0.429	12
	pH	0.21	0.498	12
	Chl *a*	0.50	0.089	12
	DOC	0.34	0.263	12
	TIC	**0.80**	**0.001**	**11**
	NH_4^+	**0.87**	**< 0.001**	**12**
	NO_3^-	−0.11	0.733	12
	TP	**0.84**	**< 0.001**	**12**
	Q	0.34	0.263	12
	v	0.27	0.389	12
	cond.	0.56	0.055	12

* Spearman Correlation was used for detecting the significance (*p*) of correlations (*r*). Bold numbers show significant correlations indicated by $p < 0.05$. *n* represents the number of compared values. Used values are means. Following further abbreviations were used: CO_2 = carbon dioxide, O_2 = oxygen, Chl *a* = chlorophyll *a*, DOC = dissolved organic carbon, TIC = total inorganic carbon, NH_4^+ = ammonium, NO_3^- = nitrate, TP = total phosphorus, *Q* = discharge; *v* = velocity; cond. = conductivity.

BGD
10, 10021–10053, 2013

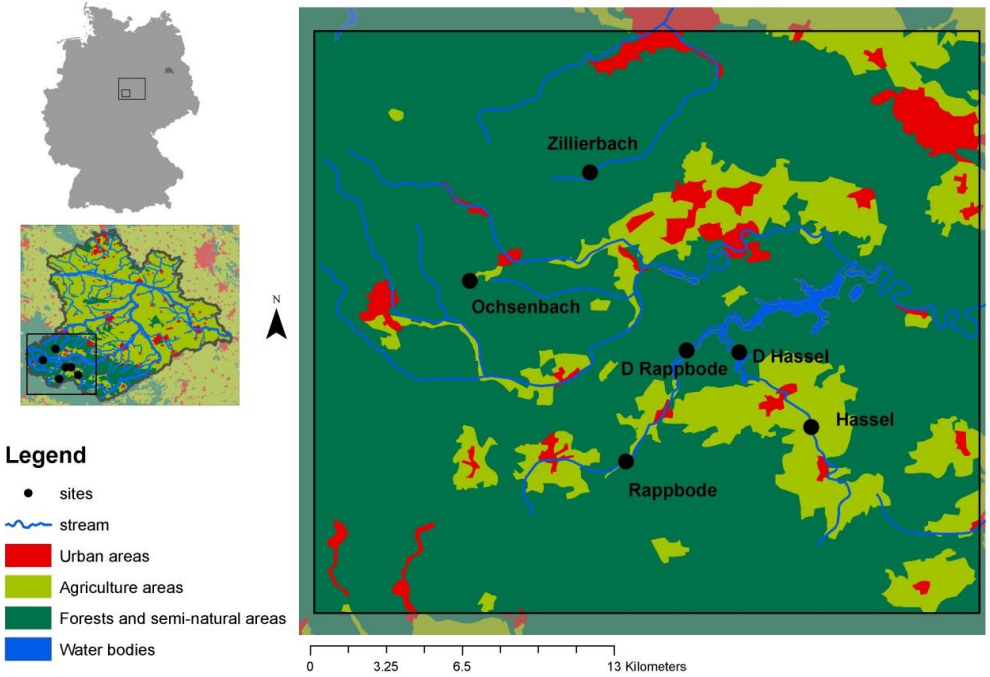

Fig. 1. Map of the investigation area. D = pre-dam.

BGD
10, 10021–10053, 2013

Different regulation of CO_2 emission from streams and lakes

S. Halbedel and M. Koschorreck

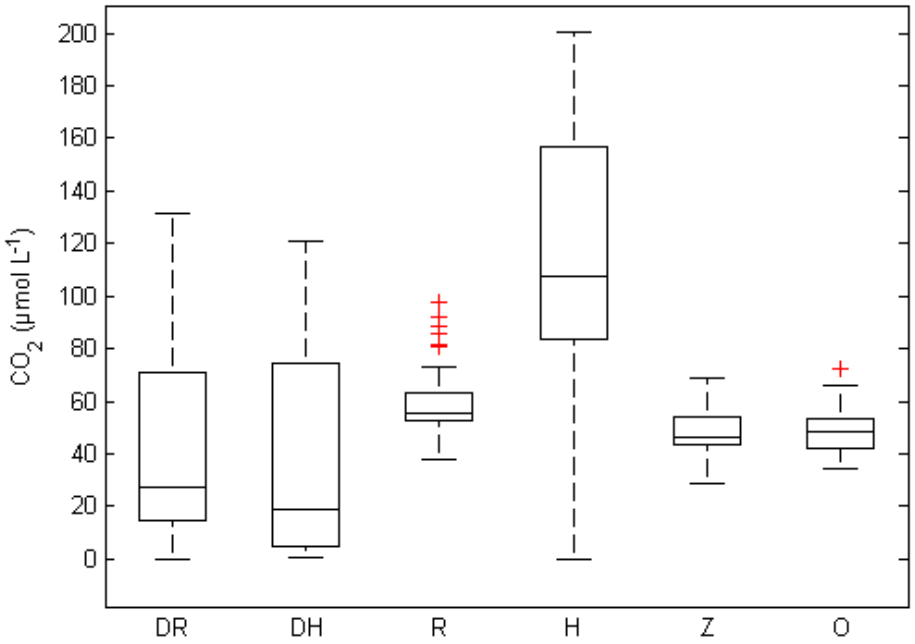

Fig. 2. Boxplots describing the average yearly CO_2 concentration (µmol L^{-1}) in the investigated streams (Rappbode (R), Hassel (H), Zillierbach (Z), Ochsenbach (O)) and pre-dams (pre-dam Rappbode (DR), pre-dam Hassel (DH)).

BGD
10, 10021–10053, 2013

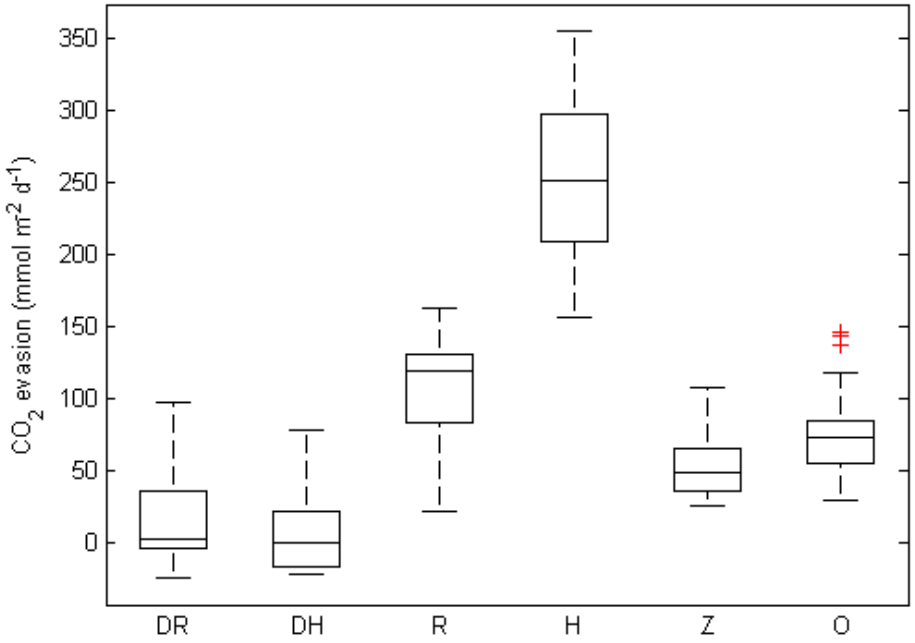

Fig. 3. Boxplots describing the average CO_2 evasion ($mmol^{-2}\,d^{-1}$) from the streams Rappbode (R), Hassel (H), Zillierbach (Z), and Ochsenbach (O) and the pre-dams Rappbode (DR) and Hassel (DH).

BGD
10, 10021–10053, 2013

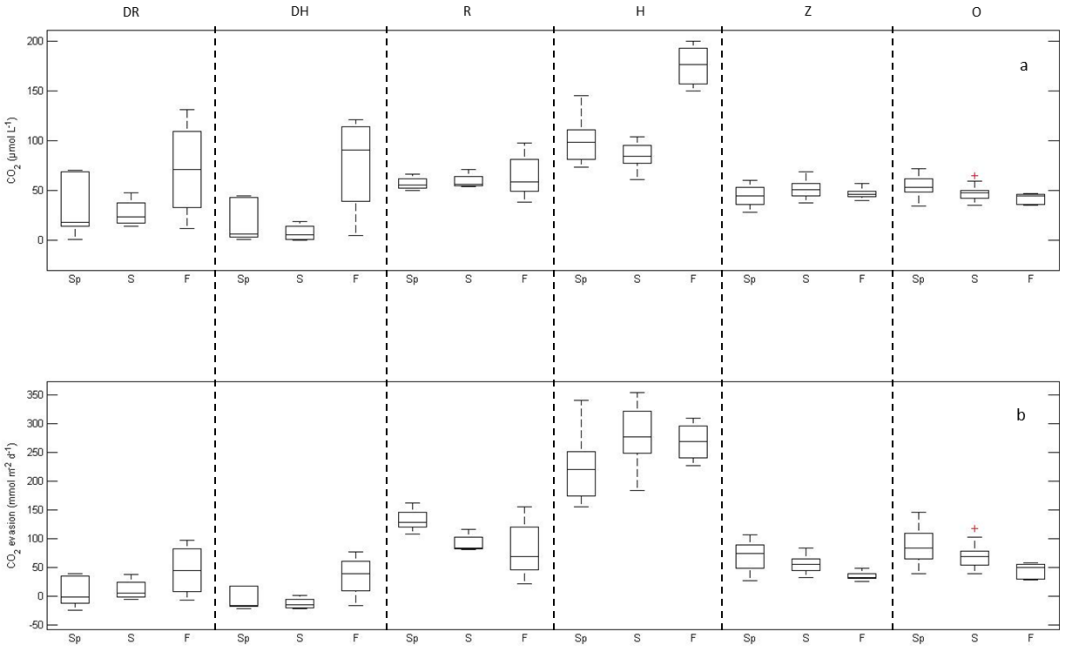

Fig. 4. Boxplots describing the annual course of CO_2 concentration **(a)** and evasion **(b)** from streams Rappbode (R), Hassel (H), Zillierbach (Z), and Ochsenbach (O) and the pre-dams Rappbode (DR) and Hassel (DH). Seasons are abbreviated as followed: Sp is spring, S is summer, and F is fall.

BGD
10, 10021–10053, 2013

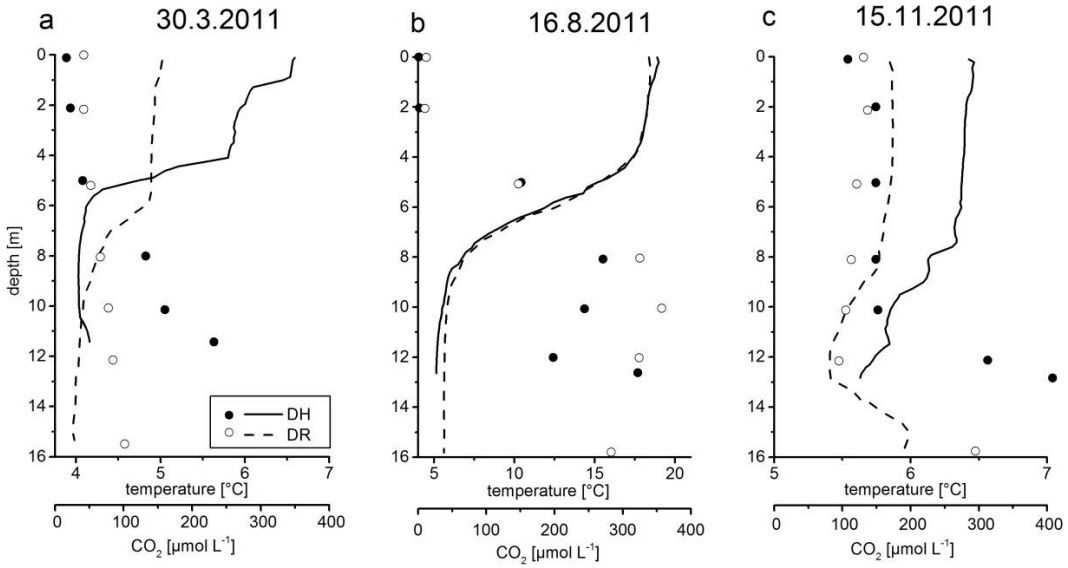

Fig. 5. Vertical profiles of temperature (lines) and CO_2 concentrations (dots) in both Pre-dams in spring **(a)**, summer **(b)**, and autumn **(c)**.

BGD
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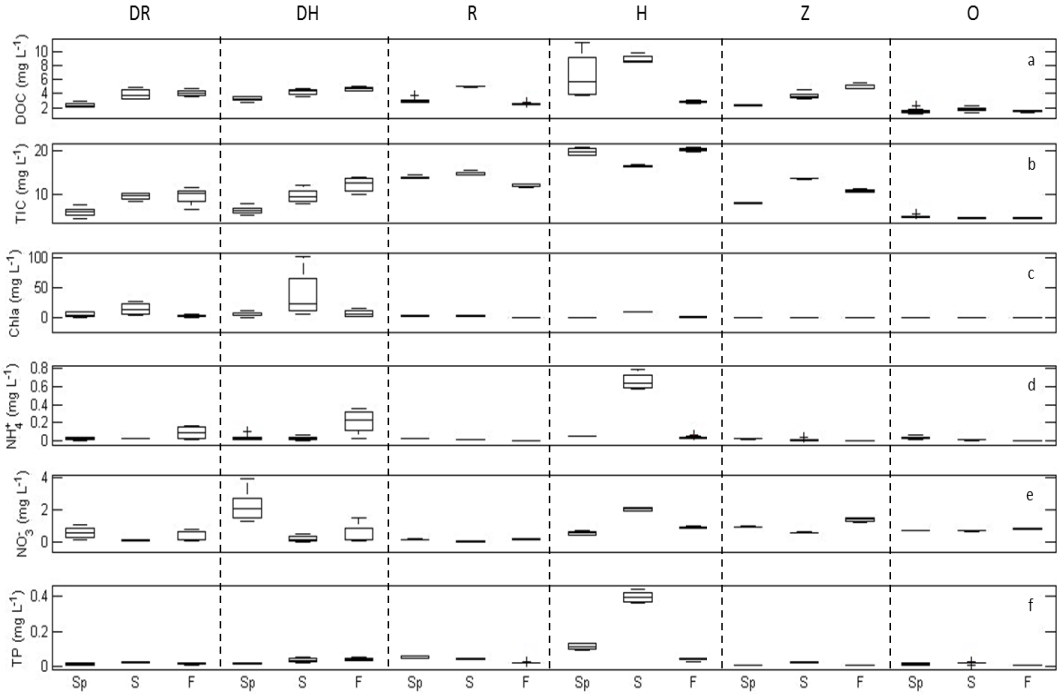

Fig. 6. Seasonal concentrations of DOC **(a)**, TIC **(b)**, Chl *a* **(c)**, NH_4^+ **(d)**, NO_3^- **(e)**, and TP **(f)** in streams Rappbode (R), Hassel (H), Zillierbach (Z), and Ochsenbach (O) and the pre-dams Rappbode (DH) and Hassel (DH). Seasons are abbreviated as followed: Sp indicates spring, S indicates summer, and F indicates fall.

BGD
10, 10021–10053, 2013

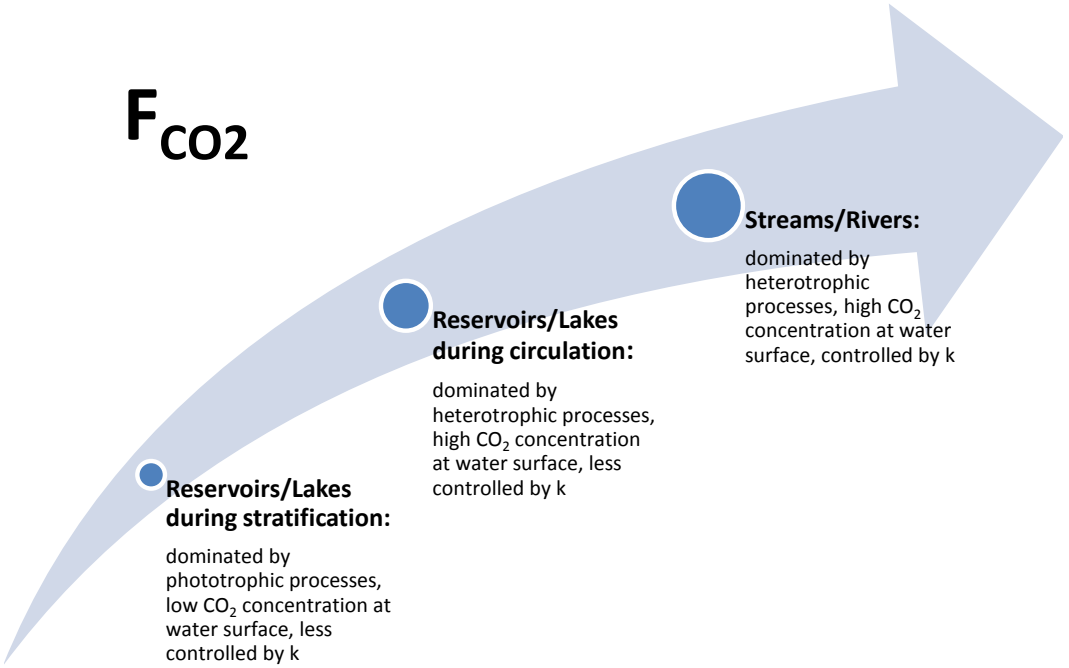

Fig. 7. Schematic presentation of the effect of different regulation mechanism on CO_2 emission (F_{CO_2}, per area) from streams and low wind lakes.

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