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Review

Temperature dependence of stream aeration coefficients and the effect of water turbulence: A critical review

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ABSTRACT

The gas transfer velocity (K_L) and related gas transfer coefficient ($k_2 = K_L A/V$, with A, area and V, volume) at the air-water interface are critical parameters in all gas flux studies such as green house gas emission, whole stream metabolism or industrial processes. So far, there is no theoretical model able to provide accurate estimation of gas transfer in streams. Hence, reaeration is often estimated with empirical equations. The gas transfer velocity need then to be corrected with a temperature coefficient $\theta = 1.0241$. Yet several studies have long reported variation in θ with temperature and 'turbulence' of water (i.e. θ is not a constant). Here we re-investigate thoroughly a key theoretical model (Dobbins model) in detail after discovering important discrepancies. We then compare it with other theoretical models derived from a wide range of hydraulic behaviours (rigid to free continuous surface water, wave and waterfalls with bubbles). The results of the Dobbins model were found to hold, at least theoretically in the light of recent advances in hydraulics, although the more comprehensive results in this study highlighted a higher degree of complexity in θ 's behaviour. According to the Dobbins model, the temperature coefficient θ , could vary from 1.005 to 1.042 within a temperature range of 0-35 °C and wide range of gas transfer velocities, i.e. 'turbulence' condition (0.005 $< K_L < 1.28$ cm min^{-1}). No other theoretical models showed any significant variability in θ with change in 'turbulence', and only modest variability in θ with change in temperature. However, the other theoretical models did not have the same temperature coefficient θ (with 1.000 < θ < 1.056 within 0–35 °C). A model integrating turbulence and bubble mediated gas transfer velocities suggested a lower temperature dependence for bubble (1.013 $<\theta<$ 1.017) than turbulence (1.023 $<\theta<$ 1.031) mediated processes. As it stands, the effect of turbulence on the temperature dependence of gas transfer at the air-water interface has still to be clarified, although many models simulate different flow conditions which may explain some of the observed discrepancies. We suggest that the temperature dependence curves produced by the Dobbins model may be used tentatively as a simple theoretical guide for streams with free surface water but not self-aerated flows encountered in whitewater rapids, cascades or weirs. Greater awareness of the different models and conditions of applications should help choosing an appropriate correction. Three case studies investigated the effect of the temperature coefficient on reaeration and stream metabolism (photosynthesis and respiration). In practice, the temperature correction may be an important parameter under constant turbulence

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conditions, but as the range in turbulence increases, the role of temperature may become negligible in determining K_L , whatever the temperature correction. The theoretical models reviewed here are also useful references to correct K_L values determined using a reference tracer gas to a second species of interest.

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1. Introduction

1.1. The importance of reaeration

Reaeration is a critical parameter in gas flux studies/modelling at the air-liquid interface both in civil engineering and industrial processes (Gulliver et al., 1990; Vogelaar et al., 2000; Huisman et al., 2004), as well as environmental studies, such as dissolved gas concentration (e.g. oxygen partial pressure/ concentration stress on animal), green house gas (GHG) emission, denitrification (open channel method), and stream metabolism (GPP, ER) - see e.g. Yongsiri et al. (2004); Schierholz et al. (2006); Battin et al. (2008); Baulch et al. (2010); Demars et al. (2011); Wallin et al. (2011). While the present study will focus on oxygen in stream, the same principles will hold for GHG (CO₂, CH₄, N₂O), N₂, and tracer gas (SF6, propane) in aquatic ecosystems (e.g. Jones and Mulholland, 1998; Huisman et al., 2004). Reaeration is also generally one of the least constrained parameter in models (e.g. Cox, 2003; Izagirre et al., 2007).

1.2. Theoretical versus empirical equations and direct measurements of reaeration coefficients

There is a plethora of theoretical models considering an interesting range of possible mechanisms but agreements among them are generally too broad (~an order of magnitude) when applied to real systems (Owens et al., 1964; Wilson and Macleod, 1974; Genereux and Hemond, 1992; Aristegi et al., 2009). The theoretical models also have at least one constant that need to be fitted with empirical data (either from laboratory or stream data), except perhaps Lamont and Scott (1970). Lamont and Scott (1970) model, while performing broadly well across aquatic habitats (Zappa et al., 2007), cannot generally represent satisfactorily the many mechanisms at play in natural systems (see Jirka et al., 2010; MacIntyre et al., 2010; Vachon et al., 2010).

So, in practice, very accurate results (10% standard error) are obtained with direct measurements (e.g. Thyssen et al., 1987; Thene and Gulliver, 1990; Genereux and Hemond, 1992; Melching, 1999). Such measurements are fairly easy and

inexpensive to carry out in small streams, but it becomes more challenging in large rivers (e.g. Richey et al., 2002). In lowland rivers, the reaeration coefficient of oxygen may also be derived indirectly from the recorded dissolved oxygen curves (Odum, 1956; Hornberger and Kelly, 1975; Chapra and Di Toro, 1991). Such indirect methods are not suitable for streams with high reaeration coefficient or/and very low biological activity (e.g. Hornberger and Kelly, 1975; Thyssen et al., 1987).

Direct measurements are also only possible for a discrete moment in time and space. The diel variability in reaeration coefficients has rarely been attempted (e.g. Tobias et al., 2009), and long term continuous estimation of reaeration coefficient need to be based on rating curves with discharge (e.g. Jones and Mulholland, 1998; Acuña et al., 2004; Roberts et al., 2007) but this is site specific and may not always work due to stream geomorphology (e.g. Genereux and Hemond, 1992; Wallin et al., 2011). The alternative use of sound in streams with standing broken waves (Morse et al., 2007) is ingenious as it relates perhaps better to stream turbulence at the air—water interface. Direct continuous measurements of turbulence at the water—air interface for gas transfer studies are at the core of several recent studies (e.g. Janzen et al., 2010; MacIntyre et al., 2010; Vachon et al., 2010).

With the need to scale up both in space, from river reach to river basin, and time, from days to years, the use of empirical relationships involving predictors such as depth, slope, and velocity (Owens et al., 1964; Moog and Jirka, 1995; Melching and Flores, 1999; Butman and Raymond, 2011; Raymond et al., 2012) have been popular. Those relationships, however, should not be used outside the predictors range and type of streams for which they were calibrated.

While the temperature dependence of the reaeration coefficients should reveal itself through the temperature variation of the physical properties involved in the theoretical models (e.g. Daniil and Gulliver, 1988; Urban and Gulliver, 2000; Gualtieri et al., 2002), empirical models of the reaeration coefficients need corrections for temperature.

1.3. Mass transfer equation and reaeration gas coefficients

The mass transfer of a slightly soluble non-reactive gas (e.g. oxygen) across an air—water interface is controlled by the liquid phase where gas molecular diffusion is about 10,000 times slower than in air and is generally written as follows:

$$\frac{\partial C_{W}}{\partial t} = k_{2}(C_{S} - C_{W}) \tag{1}$$

with C gas concentration (mg L^{-1}); C_s , saturated dissolved gas concentration (mg L^{-1}); C_w , dissolved gas concentration in the volume of water (mg L^{-1}); t, time (min) and k_2 gas exchange coefficient (min⁻¹). k_2 is determined by the product of gas transfer velocity K_L (cm min⁻¹) and specific surface area a (cm⁻¹), the latter expressed as air—water interface area (A, cm²) per unit volume of air and water (V, cm³). Hence, we have:

$$k_2 = \frac{K_L A}{V} \tag{2}$$

In stream with smooth surface water, we have $K_L = k_2 h$, with h average stream depth (cm). In self-aerated flows, measurements of specific surface area (A/V) become non-trivial due to bubbles and spray (see e.g. Toombes and Chanson, 2005; Wilhelms and Gulliver, 2005).

1.4. The temperature dependence of reaeration gas coefficients

 K_L and k_2 coefficients increase with increasing temperature, itself decreasing water viscosity and therefore facilitating the molecular diffusivity of dissolved gases (e.g. as formulated in the Stokes–Einstein equation). This rate of increase of K_L and k_2 with temperature (temperature coefficient) is the focus of the present study.

The coefficients (k_2 and K_L) are commonly temperature corrected relative to 20 °C with the simplified Arrhenius equation (see Appendix S1 in supplementary information for the full Arrhenius equation) as follows:

$$k_{\rm T} = k_{20} \theta^{(T-20)}$$
 (3)

with T, observed stream temperature; and θ , temperature coefficient, generally reported to be $\theta=1.0241$, i.e. an increase at the geometric rate of 2.41% per °C (Kilpatrick et al., 1989; Bott, 2007, p. 671; Stenstrom, 2007), no doubt based on the meticulous bottle experiments of Elmore and West (1961) but also most likely because some prominent theoretical models for free water surface confirmed those experimental results (e.g. King, 1966; Lamont and Scott, 1970; Wilson and Macleod, 1974; Gualtieri and Gualtieri, 2004). Other constant values for θ have been used, generally based on older experiments, probably unaware of the sharp criticisms by e.g. Elmore and West (1961); cf Kothandaraman and Evans (1969).

1.5. Is the temperature coefficient θ independent of turbulence?

Although Elmore and West (1961) did not find significant differences in θ with change in turbulence, both earlier studies (Kishinevsky, 1954; Kishinevsky and Serebryansky, 1956), and later studies (Dobbins, 1964; Metzger and Dobbins, 1967; Metzger, 1968) contradicted this finding both theoretically and experimentally. It is worth noting that Elmore and West (1961) two experiments were run within a narrow range of turbulence: $K_L = 0.02$ cm min⁻¹ to $0.05 < K_L < 0.12$ cm min⁻¹ (the latter depending on the size of the vortex that may have increased the water surface up to a very unlikely maximum of 2.4 times).

Limits in Dobbins (1964) new theoretical model were discussed (notably Thackston and Krenkel, 1965; Dobbins, 1965). In relation to the work of Kishinevsky, it was pointed out that Dobbins model applied for a wide range of turbulence with continuous water surface (free water surface), hence may be unable to representing gas transfer at extremely high mixing rates, as in Kishinevsky's model (Holley et al., 1970), river rapids (Hall et al., 2012), river cascades (Cirpka et al., 1993) or hydraulic structures (see below).

The work by Metzger (1968) was favourably accepted in the subsequent discussion by Rathbun and Bennett (1969)

studying the limits of the model (see also Bennett and Rathbun, 1972; cf Daniil and Gulliver, 1988), although Kothandaraman and Evans (1969) warned about potential pitfalls in the comparison of experimental data and experimental design (see also Boyle, 1974). To our knowledge, no response from Metzger followed. Only Daniil and Gulliver (1988) reported some qualitative experimental discrepancies with the Dobbins-Metzger model and concluded that the effect of turbulence intensity on the temperature correction coefficient θ had still to be clarified. Generally, the Dobbins-Metzger results have been widely accepted contrary to other heavily criticised results such as Howe (1977; cf Brown and Stenstrom, 1980; Rathbun, 1981) and Chao et al. (1987a,b; cf Daniil and Gulliver, 1989a,b; Rathbun, 1989a,b; Wilcock and McBride, 1989). Indeed the decreasing K_L with increasing temperature and $\theta < 1$ in Howe and Chao et al. studies do not seem tenable.

Early experimental work at hydraulic structures (weirs) by Gameson et al. (1958) reported a temperature dependence of 1.4–2% per °C in the range 0–35 °C, independently of fall height and pollution effects. The independence of turbulence and mixing intensity on the temperature dependence was confirmed theoretically by Gulliver et al. (1990), which also reported that Eq. (3) above (with $\theta=1.0241$) did not describe as well the temperature dependence, likely because θ was determined in a stirred container experiment without bubbles. The theoretical model used by Gulliver et al. (1990) did not include explicitly however the role of gas solubility, which has been shown to be important (Asher et al., 1997) and known to change with temperature (Battino et al., 1983).

In their textbook, Thomann and Mueller (1987) recognised that θ may generally vary from 1.005 to 1.03 based on previous specialised synthesis reports ultimately referring to the work of Dobbins and Metzger. Of course, when $\theta \rightarrow 1$, there is no need for a temperature correction (e.g. Genereux and Hemond, 1992; Demars et al., 2011). Hence, particularly for very turbulent systems $\theta = 1.0241$ has not always been readily accepted (e.g. Eheart and Park, 1989; Baulch et al., 2010; Demars et al., 2011; Wallin et al., 2011). But the logic behind this choice may be questioned since Dobbins and Metzger model was limited to free surface water and contradicting results have been presented for highly turbulent systems (e.g. Kishinevsky, 1954 versus Gulliver et al., 1990).

Rather than using $\theta = 1.0241$ indiscriminately for some work on whole stream metabolism, Demars et al. (2011) decided to calculate θ from the original formulae of Dobbins and Metzger (op. cit.). It soon appeared that while the original papers presented succinctly the theory, they did not provide all the necessary details to repeat the calculations and did not compute the temperature correction coefficient θ for a wide range of temperature and stream turbulence. Moreover, on closer inspection, while rL3 term (see below) should be constant under a given temperature (Dobbins, 1964; Metzger and Dobbins, 1967), this was not true from the values reported in the tables of the Metzger papers, hence the θ curves provided in Fig. 2 of Metzger (1968) might have been incorrect. Further, comparison between theory and empirical evidence was also biased by the selective use of data (cf Elmore and West, 1961; Downing and Truesdale, 1955; Truesdale and Van Dyke, 1958) and dubious comparability of old data (e.g.

still cited or used $\theta = 1.016$ from Streeter, 1926 and $\theta = 1.047$ from Streeter et al., 1936; cf Elmore and West, 1961; Kothandaraman and Evans, 1969).

Hence the aim of the present study was to investigate whether the temperature coefficient θ is independent of temperature and turbulence through theoretical models. More specifically, the aims are (i) to provide the necessary details and repeat the calculations of the theory presented by Dobbins for a wider range of temperature; (ii) compare the results with other representative types of theoretical models; (iii) discuss the validity of the results for gas exchange studies in the light of recent advances in hydraulics; (iv) deduce the implications for the importance of θ temperature corrections for gas transfer at the air—water interface and whole stream metabolism estimates.

2. Theory

2.1. Dobbins model

The film penetration theory for gas absorption was first developed by Dobbins (1956). The implications of the Dobbins model for the temperature coefficient θ were presented in Dobbins (1964), Metzger and Dobbins (1967) and Metzger (1968).

In this theory an interfacial film is assumed to exist in a statistical sense with its composition continuously replaced in a random manner by liquid from beneath the surface. The resulting equation is (Dobbins, 1956, 1964):

$$K_{\rm L} = \sqrt{D_{\rm m} r} {\rm coth} \sqrt{\frac{r L^2}{D_{\rm m}}} \tag{4}$$

with K_L , liquid film gas transfer velocity (cm s⁻¹),

 $D_{\rm m}$, molecular diffusivity (cm² s⁻¹),

r, average frequency of replacement of the liquid film (s⁻¹), L, film thickness (cm).

This equation reduces to the penetration model ($K_L = \sqrt{D_m r}$) for high renewal rate r, and approaches the film model ($K_L = D_m/L$) as the renewal rate, r, approaches zero (see Table 1). Hence, $K_L \propto (D_m)^n$ with 0.5 < n < 1.

The frequency of replacement of the liquid film (r) and the effective film thickness (L) were suggested to be (Metzger and Dobbins, 1967):

$$r = \frac{B\rho \nu^{3/4} E^{3/4}}{M_S} \tag{5}$$

$$L = C_4 \left(\frac{\nu^3}{E}\right)^{1/4} \tag{6}$$

with B (cm $^{-1}$), C_4 (g 4 cm $^{-10}$ s 9) $^{1/4}$ constants; ρ , water density (g cm $^{-3}$); ν , kinematic viscosity (cm 2 s $^{-1}$); E, energy dissipation in the fluid as a whole (cm 2 s $^{-3}$); and M_S, two-dimensional bulk modulus at the surface (g cm $^{-1}$ s $^{-2}$).

The parameters r and L were related, assuming M_S constant, as follows (Metzger and Dobbins, 1967):

$$r = \frac{1.3\rho r^3}{L^3} \tag{7}$$

Table 1 — Selection of theoretical reaeration coefficient (K_L) models. Refer to the original studies cited in first and last columns to find out all the assumptions and derivation of the models.								
Model	$K_L =$	With	Comment					
Film Lewis and Whitman (1924)	$\frac{D_{\mathrm{m}}}{\mathrm{L}}$							
Penetration Danckwerts (1951)	$\sqrt{D_{\mathrm{m}}r}$							
Film-penetration Dobbins (1956)	$K_L = \sqrt{D_m r} coth \sqrt{\frac{rL^2}{D_m}}$	$r = \frac{1.3\rho v^3}{L^3}$, r and L equations in text	Dobbins (1964); Metzger and Dobbins (1967)					
Friction velocity model Jähne et al. (1987a,b)	$\beta^{-1}u^*Sc^{-n}$	n ranges from 0.5 (penetration model) to 1 (film model) and β dimensionless constant (Stanton number)	Jähne et al., 1979; Jähne and Münnich, 1980					
Large eddy Wilson and Macleod (1974)	$uSc^{-1/2}Re^{-1/2}$	$Re = \frac{uA}{\nu} \approx \frac{uh}{\nu}$	O'Connor and Dobbins (1956, 1958); Fortescue and Pearson (1967)					
Small eddy Lamont and Scott (1970)	$0.4 {\rm Sc}^{-1/2} (\epsilon \nu)^{1/4} { m similar}$ to $u' {\rm Sc}^{-1/2} {\rm Re}^{-1/4}$	$\varepsilon \approx \frac{u^{\prime 3}}{\Lambda} \frac{u^{\prime} \Lambda}{v}$ Re = $\frac{u^{\prime 3}}{v}$	Banerjee et al. (1968)					
Surface divergence Banerjee and MacIntyre (2004) [0.3($0.2u^*Sc^{-1/2}Re^{-1/2}$ $2.83Re^{3/4} - 2.14Re^{2/3}$	$Re \approx \frac{u^*h}{\nu}$	Banerjee (1990)					
Turbulent thin film Prasher and Fricke (1974)	$\frac{0.0108}{\pi} \left(\frac{\rho}{\sigma}\right)^{\frac{1}{2}} \left(\frac{L^3 \epsilon}{g \nu}\right)^{\frac{1}{4}} D_m \frac{1}{2}$	$L \approx 0.3 \left(\frac{3\nu^2}{g}\right)^{\frac{1}{3}} \left(\frac{\text{Re}}{4}\right)^{\frac{8}{15}}$ $\varepsilon \approx 3.3 \left(\frac{g^4\nu}{3}\right)^{\frac{1}{3}} \left(\frac{\text{Re}}{4}\right)^{\frac{7}{15}}$	Calibrated with gas—liquid systems in falling thin films					
Films theory Gualtieri and Gualtieri (2004)	$\left(D_{\rm m}\right)^{2\!\!/_{\!3}}\!\left(\!\frac{gS}{2\nu R_{g-t}}\!\right)^{1\!\!/_{\!3}}\!=D_{\rm m}\delta_c^{-1}$	$\begin{split} R_{g-t} &= \left(\frac{\delta_{\upsilon}^{3} g S}{2 \nu^{2}}\right) \\ \delta_{\upsilon} &= \delta_{c} S c^{1/\!\!/_{3}} \end{split}$	$R_{g-t}=0.75$ calibrated on USGS river aeration data (Melching and Flores, 1999)					
Bubble mediated transfer Gulliver et al. (1990)	$\frac{\left(1-\phi^{3/2}\right)}{\left(1-\phi^{5/3}\right)^{3/4}}\frac{D_m^{3/2}u^{3/4}}{\left(L_E\nu\right)^{3/4}}$	$k_2 = K_L a$ and $a = c \frac{\phi}{\overline{d}}$	Azbel (1981) with c, dimensionless constant					
Bubble mediated transfer with gas solubility Asher et al. (1996, 1997)	$\begin{split} \left[K_M + B_C(K_T - K_M)\right] + B_CK_B &= \left[A_M \right. \\ \left. + B_C(A_T - A_M)\right]Sc^{-1/2} \\ \\ \left. + B_C\left(\frac{a_1}{\alpha} + b_1\alpha^{-m}Sc^{-n'}\right) \end{split}$	A_M , A_T proportionality constants for K_M , K_T transfer velocities	Values of constants A_M , A_T , a_1 , b_1 , m , n' taken from gas evasion experiments in cleaned freshwater					
Bubble mediated transfer Kawase and Moo-Young (1992)	$0.47D_{\rm m} \frac{1}{2} \nu^{-1/6} g^{\frac{1}{3}}$		${\rm K_L}$ almost constant regardless of bubble size and flow situation					
Turbulent diffusion Bennett and Rathbun (1972)	$\frac{2}{\sqrt{\pi}}\sqrt{D_t r}$	with $D_{\text{m}} << D_{\text{t}}$	Kishinevsky (1954); Kishinevsky and Serebryansky (1956)					

a, specific surface area $[L^{-1}]$ α , Ostwald solubility coefficient [dimensionless] B_C , fractional area bubble plume coverage [dimensionless] \overline{d} , mean bubble diameter [L] D_m , molecular diffusivity $[L^2 T^{-1}]$ D_t , turbulence diffusivity $[L^2 T^{-1}]$ δ_v , thickness of the velocity boundary layer [L] δ_c , thickness of the diffusive or concentration boundary layer [L] ϵ_c energy dissipation rate near the interface $[L^2 T^{-3}]$ η , dynamic viscosity $[M L^{-1} T^{-1}]$ g, standard gravity $[LT^{-2}]$ h, average stream or channel water depth [L] K_M , K_T , K_B transfer velocities due to turbulence of mixing pump, simulated breaking wave and $[L T^{-1}]$ bubbles respectively $[L T^{-1}]$ L, film thickness [L] L_E , size of largest eddies [L] Λ , turbulence integral length scale [L] ϕ , gas void fraction [dimensionless] r, average frequency renewal of the liquid film $[T^{-1}]$ Re, Reynolds number [dimensionless] R_{g-t} , Reynolds number for gas transfer [dimensionless] ρ , water density $[M L^{-3}]$ S, slope [dimensionless] $SC = \frac{\nu}{D_m}$, Schmidt number [dimensionless] σ , water surface tension $[M T^{-2}]$ u, average stream or channel velocity $[L T^{-1}]$ u, integral velocity scale $[L T^{-1}]$ u^* , friction velocity $[L T^{-1}]$ ν , kinematic viscosity $[L^2 T^{-1}]$.

in which the constant 1.3 (s 2 g $^{-1}$) was determined experimentally (Metzger and Dobbins, 1967; Metzger, 1968), with 1.3 equals C_5/M_S . Since we have:

$$M_{S} = c^{2} \rho \tag{8}$$

with c, speed of sound under water (cm s⁻¹), and both c and ρ are temperature dependent, the renewal rate was also computed as (Metzger and Dobbins, 1967):

$$r = \frac{C_5 \rho \nu^3}{M_5 L^3} \tag{9}$$

with C_5 (cm⁻¹) constant and M_S bulk modulus calculated from polynomial equations relating ρ and c to water temperature (Hutchinson, 1957, p. 204; Marczak, 1997). The inclusion of M_S may accommodate for the effect of surfactants (Asher and Pankow, 1991; McKenna and McGillis, 2004).

The temperature dependence of K_L rests on D_m , ρ and ν . D_m was taken as 2.14 10^{-5} cm² s⁻¹ at 20 °C and corrected to other temperature using the Stokes–Einstein equation (as in original papers) as follows:

$$D_{\rm m} = \frac{kT}{3.491\pi nr} \tag{10}$$

with k, Boltzmann's constant (1.3805 10^{-16} g cm² s⁻² K⁻¹).

- T, water temperature (K).
- η , dynamic (=absolute) viscosity of water (g cm⁻¹ s⁻¹).
- r, half the collision diameter (radius) of O_2 molecule in water (1.73 10^{-8} cm; Edward, 1970).

Note that the denominator is basically a drag coefficient. The above parameters gave excellent correspondence to the values reported in the original papers (op. cit.), although the reference value used ($D_{\rm m}$ at 20 °C) is in the upper end of the range of values determined experimentally (St-Denis and Fell, 1971; Han and Bartels, 1996).

Water density, ρ (g cm⁻³), was related to temperature (0–35 °C), using data from Hutchinson (1957, p. 204), as follows ($r^2 = 0.9999$):

$$\rho = 4.53 \cdot 10^{-8} \text{ T}^3 - 8.10 \cdot 10^{-6} \text{ T}^2 + 6.20 \cdot 10^{-5} \text{ T} + 0.99987$$
 (11)

Water kinematic (ν) and dynamic (η) viscosities are related by water density ($\nu = \eta/\rho$). The dynamic viscosity was related to temperature (0–50 °C) using data from Hodgman et al. (1961, p. 2211), as follows ($r^2 = 0.9999$):

$$\eta = -7.63 \cdot 10^{-8} \text{ T}^3 + 1.02 \cdot 10^{-5} \text{ T}^2 - 5.66 \cdot 10^{-4} \text{ T} + 0.0178 \tag{12}$$

Then, the film thickness, *L* in cm, was also related to temperature for a given turbulence, assuming *E* constant with temperature as in original papers (op. cit.):

$$L = C_L v^{\frac{3}{4}} \tag{13}$$

with $C_L = C_4/E^{1/4}$ constant $(g^{-1} \ cm^{-2} \ s^3)$ independent of temperature.

Finally, a wide range of K_L values were calculated (and converted to cm min⁻¹) over a range of temperature (0–35 °C) from a wide range of film thickness L values (0.003–0.26 cm at 20 °C), and θ values were calculated as follows:

$$\theta = \left(\frac{K_{L_{T_1}}}{K_{L_{T_2}}}\right)^{\left(\frac{1}{T_1 - T_2}\right)} \quad \text{with } T_1 \neq T_2$$
 (14)

2.2. Other theoretical models

The past 60 years of literature on reaeration models was carefully reviewed and representative models, encompassing a diverse range of hydraulic behaviours, were selected to span a wide range of turbulence and systems from which θ may be deduced for a wide range of temperature and turbulence (see Table 1). Reviews of model descriptions and mechanisms of reaeration models are already available (see e.g. Jähne and Haußecker, 1998; Gualtieri et al., 2002; Banerjee and MacIntyre, 2004; Falkovich and Sreenivasan, 2006; Gualtieri and Pulci Doria, 2008; Komori et al., 2011). Here, the reader is referred to the original publications for detailed assumptions and model derivations.

2.2.1. Continuous surface water

Lewis and Whitman (1924) film model was designed for smooth water surface and assumed diffusion through a stagnant boundary layer. This model is generally too simplistic and did not hold with experimental data in flume (e.g. Jähne and Münnich, 1980). It may only be realistic for the most sluggish (near stagnant) conditions. Danckwerts' (1951) penetration model (still a benchmark model to modern hydraulics) represented more turbulent conditions, assuming a statistical renewal of the whole viscous sublayer, as shown above with the Dobbins film-penetration model. The friction velocity model has been popularised by the meticulous work of Jähne et al. (1987a,b) who showed that it holds as long as the surface water is continuous, i.e. not at the onset of wind generated waves. This model has been used in many environmental studies, albeit sometimes with some misunderstanding of the underlying assumptions (cf Asher and Wanninkhof, 1998), contributing to discrepancies between studies; e.g. the ratio k_{CO_2} to $k_{\text{C}_3\text{H}_8}$ used varied substantially between studies: 1.3 in Jones and Mulholland (1998), 1.05 in Hope et al. (2001); 0.97 in Wallin et al. (2011). The large eddy (Fortescue and Pearson, 1967) and small eddy (Banerjee et al., 1968) models are also popular models (and some more complex models can generally be reduced to these simple models). The surface divergence model (Banerjee, 1990) was selected as it combined the large and small eddy models. The eddy models have been tested extensively in flumes (e.g. Gulliver and Halverson, 1989). A turbulent thin film (Prasher and Fricke, 1974) was also selected as a contrast to the eddy models. And finally, the films model of Gualtieri and Gualtieri (2004) was selected as it was calibrated for the USGS river aeration data (the most comprehensive and accurate river dataset available until Raymond et al., 2012).

The temperature dependence of K_L in Gualtieri and Gualtieri films model may not rest on the calibrated model with $R_{g-t}=0.75$, since it prevents R_{g-t} to vary with temperature via the viscosity of water as apparent in Eq. (25) of Gualtieri and Gualtieri (2004):

$$R_{g-t} = \frac{\delta_v^3 gS}{2\nu^2} \tag{15}$$

The thickness of the velocity boundary layer δ_v is also temperature dependent relative to the diffusive or concentration boundary layer δ_c as follows (Eq (2) in Gualtieri and Gualtieri, 2004):

$$\delta_{\rm c} = \frac{\delta_{\rm v}}{{\rm Sc}^{\frac{1}{2}_{\rm S}}} \tag{16}$$

After combining Eqs (15) and (16), we have:

$$R_{g-t} = \frac{\delta_c^3 S cgS}{2\nu^2} \tag{17}$$

Hence, instead of the semi-empirical calibrated ($R_{g-t}=0.75$) equation:

$$K_{L} = (D_{m})^{2/3} \left(\frac{gS}{2\nu R_{q-t}} \right)^{1/3}$$
 (18)

we should test the temperature dependence on the theoretical model, after substituting R_{g-t} in Eq (18) by Eq (17) to obtain:

$$K_{L} = D_{\rm m} \delta_{\rm c}^{-1} \tag{19}$$

incidentally a more simple equation showing that the temperature dependence is just a function of $D_{\rm m}$. Finally, according to Gualtieri (2005) we also have another formulation of the same model including explicitly the dissipation energy. The temperature dependence of K_L is more complicated to study and is presented as supplementary information in Appendix S2.

In order to test the effect of temperature and turbulence on the temperature coefficient θ , the K_L values of the theoretical models were calculated from a wide range of temperature (0–35 °C) and 'turbulence' parameters to get 0.005 $< K_{L20} < 1.28$ cm min⁻¹: small eddy model with energy dissipation rate $10^{-7} < \varepsilon < 10^3$ cm² s⁻³; surface divergence model with friction velocity $0.05 < u^* < 50$ cm s⁻¹ and depth h = 10 cm; turbulent thin film with $10^6 < \varepsilon < 10^7$ cm² s⁻³; films theory with slope $10^{-4} < S < 1$ for the USGS calibrated models and $0.005 < \delta_c < 0.01$ cm for the purely theoretical expression of the model.

2.2.2. Broken surface water: spray, waves and bubbles Kishinevsky (1954) turbulent diffusion model and experiments (Kishinevsky and Serebryansky, 1956) included the most extreme 'turbulent' conditions generated underwater by a blade stirrer in a cylindrical mixer. Under the most turbulent conditions, the gas diffusivity D_m became negligible to quantify K_L and $\theta=1$ whatever the temperature.

Gulliver et al. (1990) investigated the temperature dependence for gas transfer across bubble—water interface. The temperature effect upon the gas coefficients was successfully tested on Gameson et al. (1958) data, and under a given turbulence intensity, by Urban and Gulliver (2000) on the diffused aeration system of Vogelaar et al. (2000). This approach was also used for aerated tank and water reservoir with micro- and macro-bubbles Schierholz et al. (2006). The model is presented in Table 1, and we used the simplified

equation given by Gulliver et al. (1990) to compute the temperature dependence of K_L .

An additional semi-empirical model presented by Asher et al. (1996, 1997) was computed because it included explicitly gas solubility, and partitioned the overall gas transfer velocity K_L into K_M , K_T and K_B gas transfer velocities due to turbulence generated by a mixing pump, simulated breaking waves, and bubble plume, respectively (in a whitecap simulation tank). It gave the opportunity to compute K_B independently of the other transfer velocities and change the fractional area bubble plume coverage (B_C) to investigate the response of θ to changes in K_L . The temperature dependence of the Ostwald oxygen solubility coefficient was computed as in Battino et al. (1983).

An alternative theoretical model for bubble mediated gas transfer velocity was suggested by Kawase and Moo-Young (1992) and was repeatedly used in the meticulous experimental studies of Hubert Chanson quantifying the air—water interfacial area in self-aerated flows (e.g. Chanson, 1995; Chanson and Toombes, 2000; Toombes and Chanson, 2005; Felder and Chanson, 2009). A unique property of this model is that K_L is almost constant regardless of bubble size and flow situations (K_L increases with temperature from 1.8 to 3.5 cm min⁻¹ within 0–35 °C). This model is likely to apply only at cascades and hydraulic structures.

3. Results

3.1. Dobbins model

The most reliable results are produced by running the calculation of the Dobbins model either in Excel or with a small programme (c code is made available in Supplementary material, Appendix S3). Approximate values for θ may be read from the curves provided in Fig. 1 (and similar figures) or simplified equations presented in supplementary material (Appendix S4).

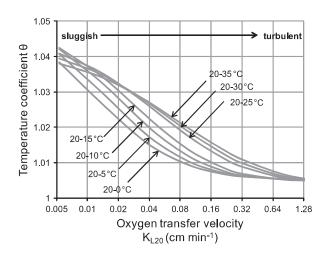


Fig. 1 — Temperature corrections derived from Dobbins' model, assuming the bulk modulus M_S constant. Newly computed values of the temperature coefficient θ relative to 20 °C against the oxygen transfer velocity K_L at 20 °C (from sluggish to highly turbulent stream flow).

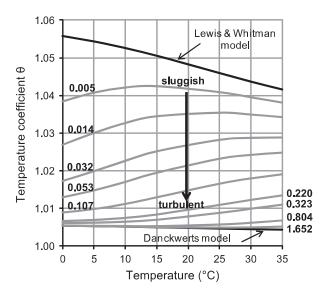


Fig. 2 — Newly computed values, derived from Dobbins' model, of the oxygen temperature coefficient θ relative to 20 °C (same as Fig. 1) against temperature, from sluggish to highly turbulent stream flow (0.005 < K_L < 1.652 values are indicated on the graph), and including the limits of the Dobbins' model: Lewis and Whitman film model (near standstill flow) and Danckwerts penetration model (highly turbulent flow).

At 20 °C, from the wide range of film thickness L values selected (0.003–0.26 cm), the average frequency of replacement of the liquid film r was 35 to 7.4 10^{-5} s⁻¹ and the resulting K_L values ranged 1.65 to 0.005 cm min⁻¹. This range represented the extreme conditions of the model. In comparison, the molecular diffusivity of oxygen (D_m) only changed from 1.1 10^{-5} to 3.1 10^{-5} cm² s⁻¹ from 0 to 35 °C, respectively.

First, assuming the bulk modulus M_S constant, θ was calculated for the temperature range 0-35 °C relative to 20 °C along a $K_{L20^{\circ}C}$ gradient of 0.005–1.28 cm min⁻¹ (Figs. 1 and 2). The temperature coefficient θ varied from 1.04 under sluggish flow conditions ($K_L = 0.005$) to about 1.005 under turbulent conditions ($K_L = 1.28$). Remarkably, the effect of temperature was qualitatively different along the turbulence gradient: θ decreased from 1.056 to 1.042 with increasing water temperature under the limit of the Lewis and Whitman film theory model; θ was then related to temperature by a 'hyperbolic' curve under sluggish flow ($K_L = 0.005$; 1.038< $\theta <$ 1.042); θ was then positively related to temperature under turbulent flow condition (e.g. $K_L = 0.05$; $\theta = 1.013-1.025$); finally under high turbulence (Danckwerts model) $\theta = 1.005$ and was not related to temperature. The temperature effect is most pronounced at intermediate turbulence ($K_L \approx 0.05$).

Then, still assuming M_S constant, θ was also calculated for a 10 °C temperature range around 5, 10, 15, 20, 25, 30 °C. The result is qualitatively very similar but quantitatively, the effect of temperature is larger (Fig. 3).

If the bulk modulus M_S is allowed to change according to temperature, the resulting changes in θ against turbulence

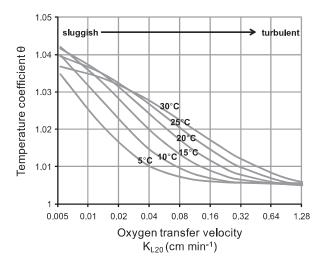


Fig. 3 — Temperature correction for empirical equations according to Dobbins' model, assuming the bulk modulus $M_{\rm S}$ constant. Same as in Fig. 1, but with temperature coefficient θ calculated for a 10 °C temperature range around 5, 10, 15, 20, 25, 30 °C and plotted against the oxygen transfer velocity $K_{\rm L}$ at 20 °C (from sluggish to highly turbulent stream flow).

and temperature are very similar to those presented above in Figs. 1–3 (see Fig. S1, S2).

3.2. Other theoretical models

The temperature coefficient θ was calculated for the temperature range 0-35 °C relative to 20 °C along a similar turbulence gradient for the additional theoretical models listed in Table 1. None of the selected models shared the property of significant (± 0.001) θ variability with turbulence as in Dobbins' model. The different models, largely representing different mechanisms akin to different systems and turbulence regimes, showed however, a temperature coefficient θ ranging from 1.000 to 1.056 within the temperature range 0-35 °C (Fig. 4). All models were sensitive to temperature, with θ negatively related to temperature, similarly to the limits of the Dobbins' model (Lewis and Whitman film model and Danckwerts penetration model) but unlike the intermediate values of the Dobbins' model (see Fig. 2). All formulations of the films models (Gualtieri) gave the same results with $\theta = 1.033 \pm 0.001$ to $\theta = 1.025 \pm 0.001$ at 0 °C–35 °C, respectively (see also Appendix S2 in Supplementary information). In the model of Asher et al. (1997), the temperature dependence of the portion of K_L due to turbulence mediated processes was higher (θ declined from 1.031 to 1.023 with increasing temperature from 0 to 35 °C) than the portion due to bubble mediated processes (θ declined from 1.017 to 1.013 with increasing temperature from 0 to 35 °C). The latter is similar to the temperature dependence of Kawase and Moo-Young (1992) model (θ declined from 1.021 to 1.016 with increasing temperature from 0 to 35 $^{\circ}$ C).

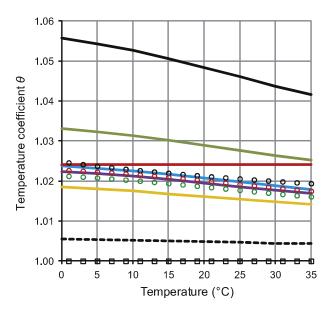


Fig. 4 — Variability in the oxygen temperature coefficient θ relative to 20 °C with temperature according to several theoretical models, see Table 1: From top to bottom, Lewis and Whitman 1924 (black line), Gualtieri and Gualtieri 2004 (green line), Urban and Gulliver 2000 (black circles), Lamont and Scott 1970 (blue line), Asher et al., 1997 (red circles), Banerjee 1990 (purple line), Kawase and Moo-Young 1992 (green circles), Prasher and Fricke 1974 (yellow line), Danckwerts 1951 (dashed black line), and Kishinevsky 1954 (black squares). The Elmore and West 1961 constant ($\theta=1.0241$) is in red for comparison. Compare to Fig. 2.

4. Discussion

4.1. How realistic is the Dobbins model?

It is interesting to note that, with the selected range of film thickness, L (0.003–0.26 cm) was comparable to direct experimental verifications (0.003 < L < 0.08 cm; e.g. Jähne et al., 1989; Asher and Pankow, 1991; Chu and Jirka, 1992; Moog and Jirka, 1999; Herlina and Jirka, 2004) and comparable to other model results (e.g. Gualtieri and Gualtieri, 2004). While such estimates were already known in the 1960s (with 0.004 < L < 0.04 cm, see Metzger and Dobbins, 1967), there was a dearth of data regarding the average frequency of replacement of the liquid film r. More recent experimental values for r validate the highest renewal rates of the liquid film (e.g. 0.1 < r < 10 s⁻¹ in Jähne et al., 1989; 0.1 < r < 100 s⁻¹ for 0.03 < K_L < 0.6 cm min⁻¹ in Komori et al., 1993).

The very gentle and rapid (distortion of the surface water) mixing conditions of Metzger and Dobbins (1967) experiments are represented by $K_L = 0.03$ and 0.3 cm min⁻¹; with L = 0.05 and 0.01 cm; and r = 0.008 and 1.3 s⁻¹ respectively.

While many authors have emphasised the role of D_m (e.g. Thackston and Krenkel, 1965; Taricska et al., 2009), the most sensitive parameter to model K_L with the Dobbins' model is r, as noted earlier (Dobbins, 1965). This sensitivity had arisen

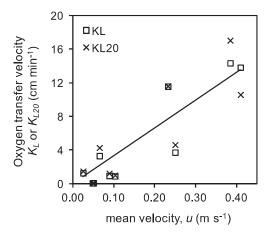


Fig. 5 – Observed (K_L) and standardised (K_{L20} with $\theta=1.024$) oxygen transfer velocities as a function of stream velocity (turbulence) in Fuirosos stream, Spain (Acuña et al., 2004).

a criticism by Holley et al. (1970) regarding the experimental results of Dobbins (1964): "It is true that r should increase as [the speed of oscillation of the metal grid] σ increases, but it is difficult to conceive that increasing σ by a factor of about 11 would have changed the characteristics of the mixing to such an extent that r would be increased by a factor greater than 20,000". So while the high average renewal rate r have been verified under experimental conditions (see above), further experimental determinations of r are required under low K_L .

Dobbins (1956) highlighted that his model could only be applied to cases in which the age distribution function of r, the same exponential function as in Danckwerts (1951), is correctly defined. Several studies have shown that the Kolmogorov distribution function (a log-normal distribution) was more appropriate (e.g. Garbe et al., 2004; Kermani and Shen, 2009). This said, the *average* surface age from Dankckwerts' random surface renewal model was surprisingly close to hybrid Lagrangian tracing and temperature method, the state of the art in gas transfer modelling at the interface between a liquid and the atmosphere (Kermani et al., 2011).

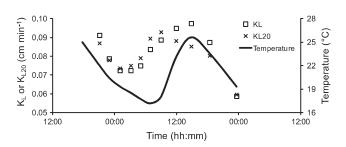


Fig. 6 – Diel change in observed (K_L) and standardised (K_{L20} with $\theta=1.024$) oxygen transfer velocities in Sugar Creek in western Indiana, USA (K_L and temperature data taken from Tobias et al., 2009).

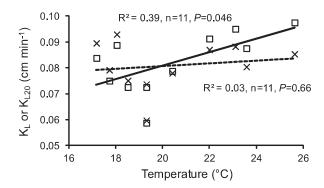


Fig. 7 — Temperature was a relatively weak predictor of K_L (black line) in Sugar Creek, western Indiana, USA (K_L and temperature data taken from Tobias et al., 2009). As expected K_{L20} was not related to temperature (dashed line). Regression equations were tested with 1000 random cyclic shift Monte Carlo permutations to take into account the temporal structure of the data.

4.2. Comparison with other models

All theoretical models, as noted in introduction, needed to be fitted onto experimental (or field) data to obtain the necessary constants. Even the small eddy model of Lamont and Scott (1970) relied on the use of constants in the energy spectrum function (their Eq. (12)) although these constants were then carried through the subsequent derivations. Moreover, applying these theoretical models request further approximations, e.g. for Re (see the eddy models in Table 1). Hence, caution should be applied when comparing the results among the theoretical models and with experimental data, particularly old data subject to potential bias (including Elmore and West, 1961; cf Kothandaraman and Evans, 1969; Boyle, 1974). This said, while the absolute values of K_L derived from theoretical models depend on empirically derived constants, the relative change in K_L due to temperature (K_{LT1}/K_{LT2}) generally does not (Gulliver et al., 1990).

Daniil and Gulliver (1988) correctly reported inadequacy between Dobbins' model and some old empirical data regarding the role of turbulence, but they only plotted the limits of the model, which provided a nice envelop around all available data point, exactly what you would expect the model to do, should it be comparable to the experimental data points (see Fig. S3). Daniil and Gulliver (1988) did note, however, discrepancies between the Dobbins model and Jähne et al., 1987a experimental results under varying turbulence.

Finally, both Jähne et al., 1987a and Gulliver and Halverson (1989) pointed to the great danger in extrapolating laboratory experimental results to field conditions. Even the eddy models incorporating Sc and Re are generally rather simplistic and are unlikely to deliver accurate predictions under field conditions because of the numerous mechanisms driving the energy dissipation rate near the interface such as wind shear, natural convection, rain, entrapped air in broken standing waves, entrained air in bubbles, spray (e.g. Chanson and Toombes, 2002; Banerjee and MacIntyre, 2004; Wilhelms and Gulliver, 2005; Vachon et al., 2010).

5. Case studies

Here we report three case studies to illustrate the importance of the temperature correction θ .

5.1. Oxygen transfer velocity rating curve

In order to estimate whole stream metabolism continuously over time, Acuña et al. (2004) carried out 10 propane tracer studies in the same stream reach at different time of year and discharge to produce a rating curve (see Fig. 5). K_L was highly related to stream mean water velocity u ($K_L = 33u$; $r^2 = 0.90$, n = 10, P < 0.001). The standardisation of K_L at 20 °C (K_{L20}) based on $\theta = 1.024$ did not improve the relationship despite a wide range of stream water temperature (9–31 °C). This may be due to the very high oxygen transfer velocity K_L , (up to >10 cm min⁻¹) which may best correspond to the turbulent model of Kishinevsky, in which case the temperature dependence is negligible.

5.2. Diel change in oxygen transfer velocity

Tobias et al. (2009) injected a saturated solution of SF6 continuously over 24 h to a stream in western Indiana, USA. Here we present the data in a different way to better illustrate the role of temperature (17–25 °C) independently of the wind (which was measured 40 km away from the field site and thus may not be very accurate), see Fig. 6. If temperature had been a strong control on the diel variability, then K_{L20} (with $\theta=1.024$) should not have shown any diel changes. It is obvious that temperature had a limited role in this case study, even though temperature could explain 38% of the variability in K_L , but, as expected, not K_{L20} (Fig. 7). Turbulence created by wind was the most likely reason of the diel variability in K_L

Table 2 — Potential discrepancy in daily stream metabolism due to water temperature fluctuation (with $\theta=1.024$) for sites with large diel temperature variability in Hengill streams, Iceland (Demars et al., 2011). The discrepancy was <2.5% at all sites using the Dobbins—Metzger model with $\theta=1.005$ (Demars et al., 2011). Site 1 is illustrated in Fig. 8.

Hengill	Temperature	Depth	k_2	K_L	-ER	-ER	-ER	GPP	GPP	GPP
Sites	Range °C	cm	$\overline{\mathrm{min}^{-1}}$	cm min ⁻¹	$\overline{\text{g O}_2 \text{ m}^{-2} \text{ day}^{-1}}$	$\delta x/x$	$\overline{\theta} = 1.024$	$\overline{g O_2 m^{-2} day^{-1}}$	$\delta x/x$	$\overline{ heta}=1.024$
1	16–23	14	0.077	1.078	28.2	38%	-11%	20.3	1%	-9%
4	7 — 15	3	0.269	0.807	2.5	85%	-8%	2.2	7%	-4%
11	11–17	3	0.506	1.518	9.7	86%	-5%	9.2	19%	-1%

 $\delta x/x$, relative uncertainty based on 1 standard deviation $\theta=1.024$, potential discrepancy due to temperature correction (Elmore and West, 1961).

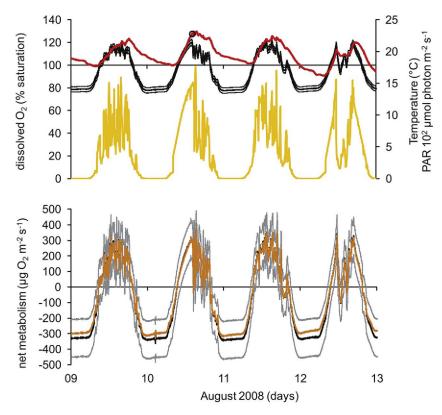


Fig. 8 – Top panel shows diel change in dissolved oxygen, temperature and photosynthetic active radiation (PAR) at site 1 in Hengill, Iceland (Demars et al., 2011); the black circle on the temperature curve indicates when the propane gas tracer study was conducted. Bottom panel shows the net metabolism of the stream without (black line with grey lines showing uncertainties) and with (orange line) temperature correction (with $\theta = 1.024$). The effect of temperature correction on daily metabolism is presented in Table 2.

(see Tobias et al., 2009). Note that here the Dobbins model gave similar results because here $\theta=1.018$ was similar to $\theta=1.024$.

5.3. Temperature correction and whole stream metabolism

Field ecologists often cannot run a continuous tracer study as Tobias et al. (2009) did, while measuring dissolved oxygen in stream, for logistical reasons. Generally the gas tracer study only last the time it takes for a slug injection of solute conservative tracer to completely pass through the studied reach. The tracer study is generally done during the day for a relatively short time (30 min to an hour, but may be several hours if water transient storage is important or the reach very long). At sites where there is a strong diel variability, it is common usage to correct the reaeration coefficient for temperature, but this may depend on the value of θ . For example, Demars et al. (2011) used the Dobbins model to compute θ and calculated that it made virtually no change in the final daily stream metabolism estimate (<2.5%). Should they have chosen $\theta = 1.024$, the error in daily metabolism at the sites with strong diel change in temperature would be larger (Table 2, Fig. 8), but by no means as large as the uncertainties computed by Tobias et al. (2009) mostly due to change in turbulence (wind velocity).

6. Conclusion

According to Dobbins theoretical model, the gas transfer velocity response to variation in temperature, which affects water properties and molecular diffusivity, is not important where turbulence is rapidly renewing the concentration boundary layer at the air-water interface. No other theoretical models showed any significant interaction effects between temperature and turbulence. However, the other theoretical models differed widely in their response to temperature. Several issues (e.g. better representation of the different field turbulence mechanisms, minimisation of the role of fitted constants, use of a more appropriate age distribution function of r) need to be addressed to develop better theoretical models for which the effect of temperature may be expressed through the physical parameters. Greater awareness of the diversity of models and conditions of applications should help choosing an appropriate correction. In practice, the temperature correction is an important parameter under constant turbulence conditions, but as the range in turbulence increases, the role of temperature may become negligible in determining K_L , whatever the correction. The theoretical models reviewed here are also useful references to correct K_L values determined using a reference tracer gas to a second species of interest or dual gaseous tracer experiments.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.watres.2012.09.054

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