Measurement and Prediction of Oxygen Transfer in Activated Sludge based on Ex Situ Off-gas Monitoring

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Abstract

This dissertation examines oxygen transfer dynamics of activated sludge aeration systems in wastewater treatment plants (WWTP). The method of ex situ off-gas testing and its measurement uncertainty when determining the α-factor were studied. The variation of the α-factor was measured in conventional activated sludge (CAS) and two-stage systems with pilot-scale long-term ex situ off-gas testing. A data-driven approach to predict oxygen transfer based on supervised machine learning is presented. The key results of this cumulative dissertation and its three papers (P1-P3) are as follows:

- ASCE 18-18 describes the ex situ off-gas method as an alternative to in situ off-gas testing with off-gas hoods on the activated sludge surface. P1 showed that results from ex situ and in situ tests cannot be compared because sludge inflow into an ex situ bubble column systematically increased the α-factor. Still, ex situ off-gas testing offers unique advantages for piloting and research of oxygen transfer because operation of an external bubble column is more flexible than in situ off-gas testing.
- By comparing ex situ off-gas measurements under the same conditions, P1 demonstrated that α-factors can be quantified at a relative standard deviation of about ± 2.8 %. This is significantly more accurate than previously reported uncertainties between ± 5 to 15 %. A sensitivity analysis in P1 revealed that recording the oxygen concentration in the off-gas was the most important parameter to conduct reliable oxygen transfer tests, exceeding the relevance of dissolved oxygen (DO) and airflow rate measurement by far.
- α-factors are generally higher in the second stage of a two-stage WWTP because oxygen transfer inhibiting substances, e.g., surfactants and TOC, are partially removed in the first stage. In P2, α-factors for design load cases were determined as 0.45 for α_{mean} and 0.33/0.54 for $\alpha_{min}/\alpha_{max}$ in the first stage (HRAS), and as 0.80 for α_{mean} and 0.69/0.91 for $\alpha_{min}/\alpha_{max}$ in the second stage. α-factors in situ would be lower because these values were recorded with ex situ off-gas tests.
- The α_0-factor was introduced in P3 to compare oxygen transfer in activated sludge from aerated and non-aerated zones. It considers differences of in situ and ex situ DO under non-steady state DO conditions. An increase of the α_0-factor along an upstream anoxic tank of a CAS process was observed, thus suggesting biosorption and/or biodegradation of oxygen transfer inhibiting substances.
- The α_0-factor was predicted by Random Forest models for different activated sludge stages within an RMSE (root-mean-square error) of 0.024 and 0.033 (R^2 between 0.84 and 0.92). Models were trained with 17 predictor variables based on WWTP operating data. The data-driven approach can consider potential interactions of influences on oxygen transfer, but the final models are typically unable to generalize for conditions not included in training data.

Kurzfassung

Im Rahmen dieser Dissertation wird der Sauerstoffeintrag von Belüftungssystemen im Belebtschlammverfahren von Abwasserbehandlungsanlagen (ABA) untersucht. Dabei wird die ex situ Abluftmessung und deren Messunsicherheit bei der Bestimmung des α-Wertes betrachtet. Mit ex situ Abluftmessungen im halbtechnischen Maßstab wurden Schwankungen des α-Wertes im Betrieb konventioneller und zweistufiger Abwasserbehandlungsanlagen aufgezeichnet. Die datengetriebene Vorhersage des Sauerstoffeintrags basierend auf Modellen des überwachten maschinellen Lernens wird aufgezeigt. Die wesentlichen Ergebnisse der kumulativen Dissertation und der drei Publikationen (P1-P3) sind wie folgt:

- In ASCE 18-18 wird die ex situ Abluftmethode als Alternative zu in situ Messungen mit Ablufthauben auf der Oberfläche von Belebungsbecken beschrieben. In P1 wurde aufgezeigt, dass Messergebnisse von ex situ und in situ Abluftmessungen nicht vergleichbar sind, da die Schlammzufuhr in eine externe Blasensäule den Sauerstoffeintrag, und damit den α-Wert, systematisch erhöhte. Dennoch bietet die ex situ Abluftmethode Vorteile für Pilotierungs- und Forschungszwecken, da diese flexibler betrieben werden kann als in situ Ablufthauben.
- Ein Direktvergleich von ex situ Abluftmessungen in P1 zeigte, dass α-Werte mit einer relativen Standardabweichung von etwa ± 2,8 % gemessen werden konnten. Diese Abweichung ist deutlich geringer als bisher bekannte Abweichungen zwischen ± 5 und 15 %. Anhand einer Sensitivitätsanalyse konnte die Messung der Sauerstoffkonzentration in der Abluft als wichtigste Einflussgröße für zuverlässige Abluftmessungen identifiziert werden. Diese war wesentlich entscheidender als die Messung der Gelöst-Sauerstoffkonzentration und des Luftvolumenstroms.
- In zweistufigen Belebungsbecken sind α-Werte in der zweiten Stufe höher, da in der Hochlaststufe Abwasserinhaltsstoffe, z.B. Tenside und TOC, teilweise entfernt werden, die den Sauerstoffeintrag hemmen. In P2 wurden α-Werte für Lastfälle bestimmt, darunter 0,45 für α_{mittel} und 0,33/0,54 für $\alpha_{min}/\alpha_{max}$ in der Hochlaststufe sowie 0,80 für α_{mittel} und 0,69/0,91 für $\alpha_{min}/\alpha_{max}$ in der zweiten Stufe. α-Werte im Belebungsbecken wären tendenziell niedriger, da diese Werte mit ex situ Abluftmessungen erhoben wurden.
- Der α_0-Wert wurde in P3 eingeführt, um den Sauerstoffeintrag in Belebtschlamm aus belüfteten und unbelüfteten Beckenzonen zu untersuchen. Der Parameter berücksichtigt den Unterschied zwischen in situ und ex situ Sauerstoffkonzentration unter stationären und instationären Bedingungen. In einer konventionellen Belebungsanlage konnte ein Anstieg des α_0-Wertes entlang der Beckenlänge einer vorgeschalteten anoxischen Beckenzone beobachtet werden. Dies weist auf einen

Abbau und/oder eine Adsorption von Abwasserinhaltsstoffen hin, die den Sauerstoffeintrag hemmen.

- Der α_0-Wert konnte mit Random Forest Modellen für verschiedene Belebtschlamm-Stufen mit einem RMSE (root-mean-square error) zwischen 0,024 und 0,033 (R^2 zwischen 0,84 und 0,92) vorhergesagt werden. Die Modelle wurden mit 17 Vorhersagevariablen aus Betriebsdaten der ABA trainiert. Der datengetriebene Ansatz kann mögliche Interaktionen zwischen Einflüssen auf den Sauerstoffeintrag berücksichtigen. Allerdings können Modelle im Allgemeinen keine zuverlässige Vorhersage für neue Bedingungen treffen, die nicht Bestandteil des Trainings-Datensatzes waren.

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State of the Art – Aeration Technology

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List of Abbreviations

Abbreviation	Unit	Description
a	$m^2 \cdot m^{-3}$	Interfacial area
AFR	$Nm^3 \cdot m^{-3} \cdot h^{-1}$	Volume specific airflow rate
AOTR	$kg \cdot h^{-1}$	Actual oxygen transfer rate under process conditions
AS	-	Activated sludge
ASCE	-	American Society of Civil Engineers
ASM	-	Activated sludge model
ASP	-	Activated sludge parameter
BOD	$mg \cdot L^{-1}$	Biological oxygen demand
CAS	-	Conventional activated sludge system
CLR	-	Closed-loop reactor
COD	$mg \cdot L^{-1}$	Chemical oxygen demand
$CO_{2,e}$	%	CO_2 concentration in off-gas
$CO_{2,in}$	%	CO_2 concentration in inlet gas
$C_{S,md}$	$mg \cdot L^{-1}$	Oxygen saturation concentration at mid-depth of tank and standard conditions
$C_{S,T,St}$	$mg \cdot L^{-1}$	Oxygen saturation concentration at water temperature T_w
CSTR	-	Continuous stirred tank reactor
C_t	$mg \cdot L^{-1}$	Dissolved oxygen concentration at time t
cw	-	Clean water (in index of oxygen transfer parameters)
C_0	$mg \cdot L^{-1}$	Dissolved oxygen concentration at start of test
C^*_{20}	$mg \cdot L^{-1}$	Standardized effective oxygen saturation at process conditions
C^*_{∞}	$mg \cdot L^{-1}$	Steady-state dissolved oxygen saturation concentration as time approaches infinity
DN	-	Denitrification zone
DO	$mg \cdot L^{-1}$	Dissolved oxygen (concentration)

DO_{zone}	$mg \cdot L^{-1}$	Dissolved oxygen (concentration) in external activated sludge tank zone during ex situ testing
DOC	$mg \cdot L^{-1}$	Dissolved organic carbon
DWA	-	Deutsche Vereinigung für Wasserwirtschaft, Abwasser und Abfall e. V.
DWP	-	Dynamic wet pressure
EC	$\mu S \cdot cm^{-1}$	Electrical conductivity
EN	-	European Standard, European Norm
EP	-	Effluent parameter
EPA	-	U.S. Environmental Protection Agency
EPS	-	Extracellular polymeric substances
F	$m^3 \cdot h^{-1}$	Flow
F	-	Fouling factor
F/M ratio	$kg \cdot kg^{-1} \cdot d^{-1}$	Feed to mass ratio or food to microorganism ratio
h_D	m	Blow-in depth
HFV	%	Hydrostatic floc volume
HRAS	-	High-rate activated sludge system
HRT	min	Hydraulic retention time
HRT_a	min	Actual hydraulic retention time
HRT_n	min	Nominal hydraulic retention time
IP	-	Influent parameter
k_L	$m \cdot s^{-1}$	Liquid film coefficient
k_La	h^{-1}	Volumetric oxygen mass transfer coefficient
k_La_{20} or $k_La_{20,cw}$	h^{-1}	Volumetric oxygen mass transfer coefficient at 20 °C in clean water
$k_La_{20,1000}$	h^{-1}	Volumetric oxygen mass transfer coefficient at 20 °C and 1,000 $mg \cdot L^{-1}$ TDS
KNN	-	k-nearest neighbors
L × W × H	m	Length × Width × Height (reactor dimensions)
MAE	-	Mean absolute error
MBR	-	Membrane bioreactor

MCRT	d	Mean cell retention time
ML	-	Machine learning
MLSS	$g \cdot L^{-1}$	Mixed liquor suspended solids
MLVSS	$g \cdot L^{-1}$	Mixed liquor volatile suspended solids
MR_e	-	Molar ratio of oxygen in the inlet gas
MR_i	-	Molar ratio of oxygen in the off-gas
n	-	Number of observations in a dataset
$\mathcal{N}$	-	Normal distribution
NDIR	-	Non-dispersive infrared (gas analyzer)
OAT	-	"One factor at a time" analysis
OTE	%	Oxygen transfer efficiency
OTE_{pw} or OTE_f	%	Oxygen transfer efficiency under process conditions (process water), OTE_f commonly used in ASCE publications
$OTE_{sp,20}$	$\%/(mg \cdot L^{-1})$	Oxygen transfer efficiency per unit of driving force at std. conditions
OTR	$g \cdot h^{-1}$	Oxygen transfer rate
OUR	$g \cdot h^{-1}$	Oxygen uptake rate by the microorganisms
$O_{2,e}$	%	O_2 concentration in off-gas
$O_{2,in}$	%	O_2 concentration in inlet gas
P	hPa	Pressure
P1, P2 or P3	-	Paper 1, Paper 2 or Paper 3
p_{atm}	hPa	Atmospheric pressure
PCA	-	Principal component analysis
PC1 or PC2	-	Principal component 1 or 2
PE	-	Population equivalent
PFR	-	Plug flow reactor
pw	-	Process water (in index of oxygen transfer parameters)
Q	$m^3 \cdot h^{-1}$	Flow
Q1	-	First quartile (25 percentile)

Q3	-	Third quartile (75 percentile)
Q_{AFR}	$Nm^3 \cdot h^{-1}$	Airflow rate, usually at standard temperature and pressure
q_{air}	$m^3 \cdot h^{-1}$	Airflow rate
$q_{air,vol}$ or $q_{Vol,aer}$	$Nm^3 \cdot m^{-3} \cdot h^{-1}$	Volume specific airflow rate
Q_{AS}	$m^3 \cdot h^{-1}$	Activated sludge flow into ex situ reactor
RAS	-	Return activated sludge
RF	-	Random Forest
RFE	-	Recursive feature elimination
RMSE	-	Root-mean-square error
R^2	-	Coefficient of determination
SA	-	Sensitivity analysis
SAE	$kg \cdot kWh^{-1}$	Standard aeration efficiency
SBR	-	Sequencing batch reactor
SD	-	Standard deviation
SNR	-	Signal to noise ratio
$SOTE_{cw}$	%	Standard oxygen transfer efficiency under test conditions (clean water)
$SOTE_{pw}$	%	Standard oxygen transfer efficiency under process conditions (process water)
$SOTR_{cw}$	$kg \cdot h^{-1}$	Standard oxygen transfer rate in clean water
$SOTR_{pw}$	$kg \cdot h^{-1}$	Standard oxygen transfer rate in process water
SRT	d	Sludge retention time
SVI	$mL \cdot g^{-1}$	Sludge volume index
TDS	$mg \cdot L^{-1}$	Total dissolved solids
TOC	$mg \cdot L^{-1}$	Total organic carbon
TOC_{in}	$mg \cdot L^{-1}$	Total organic carbon in inflow
TS	$g \cdot kg^{-1}$	Total solids
TSS	$g \cdot L^{-1}$	Total suspended solids
$TSS_{effluent}$	$g \cdot L^{-1}$	Total suspended solids in effluent
T or T_w	°C	Wastewater temperature (in activated sludge)

V	m^3	Volume
WAS	-	Waste activated sludge
WWTP	-	Wastewater treatment plant
α	-	Alpha-factor – correction factor for oxygen transfer parameters in process water
α_0	-	Alpha(zero)-factor – correction factor for oxygen transfer parameters in process water under non-steady state conditions
β	-	Beta-factor – wastewater correction factor for oxygen saturation
μ	-	Mean value
σ	-	Standard deviation
σ^2	-	Variance
θ	-	Theta-factor – temperature correction factor
τ	-	Temperature correction (tau) of effective saturation concentration
Ω	-	Omega-factor – pressure correction factor

1 Introduction

Aeration of activated sludge is an essential process in biological wastewater treatment where aeration systems supply oxygen to aerobic microorganisms. This transfer of oxygen from gas into liquid phase is an energy-intensive process. In the last decades major advances were put into practice to improve energy efficiency of this process, e.g., the shift from surface to submerged aeration systems, from coarse to fine-bubble diffusers, and automation of dissolved oxygen control systems (Wagner and Stenstrom, 2014). Nonetheless, aeration still accounts for more than half of the net energy consumption in most wastewater treatment plants (WWTP) (Baquero-Rodríguez et al., 2018; Reardon, 1995; Rosso et al., 2011). Hence, even minor improvements of aeration efficiency can translate to significant cost savings for WWTP operators. This cumulative dissertation addresses research gaps in the field of aeration technology in three research papers as described below.

Paper 1 (P1) addresses missing information about oxygen transfer testing procedures for the ex situ off-gas method that was used throughout this research work. Technical guidelines to quantify the oxygen transfer efficiency in clean water (ASCE/EWRI 2-06, 2007; DWA-M 209, 2007; EN 12255-15, 2003) and process water (ASCE/EWRI 18-18, 2018; DWA-M 209, 2007) exist to standardize measurement procedures. These allow to compare the efficiency of aeration systems and WWTP process layouts and are a critical tool for the development and design of aeration systems to improve aeration efficiency. However, oxygen transfer tests under process conditions are reported to produce results within an uncertainty of ± 5 to 15 % if conducted according to testing procedures (Capela et al., 2004; DWA, 2007; Mahendraker et al., 2005; Redmon et al., 1983). Therefore, in P1 the parameters critical for measurement uncertainty are examined and discussed. An important oxygen transfer parameter is the α-factor that describes the ratio of oxygen transfer under process and clean water conditions. The α-factor is determined by in situ or ex situ off-gas procedures based on a mass balance of oxygen transferred from gas to liquid phase. However, application experience with ex situ off-gas methods is limited. Hence, P1 discusses applications and limitations of the ex situ off-gas method.

Paper 2 (P2) focusses on the oxygen transfer in two-stage activated sludge treatment plants. In this process layout a high-rate activated sludge system (HRAS) is followed by a second biological treatment stage (e.g., for nitrogen removal), that can be operated differently than a conventional activated sludge (CAS) system (Winkler and Widmann, 1994). A key difference is the much lower wastewater load directed into the second stage after HRAS treatment. Recently, two-stage processes gained attention because the first stage enables the redirection of carbon into waste activated sludge (WAS) through biosorption, thus reducing the oxygen demand of aerobic biodegradation. While almost all CAS wastewater treatment plants operate in an energy-negative mode, two-stage systems were reported to energy self-sufficiently remove nutrients (Kroiss and Klager, 2018; Liu et al., 2020). Because two-stage systems are operated differently than CAS systems this also affects oxygen transfer. A lower α-factor due to high sludge loads is expected in HRAS systems followed by higher α-factors in the subsequent treatment stage. However, no comprehensive information about two-stage process and oxygen transfer is available yet. Therefore, P2 examines effects on oxygen transfer during operation in practice and defines static α-factors for design purposes based on long-term ex situ off-gas testing.

Paper 3 (P3) presents a novel approach to model oxygen transfer in activated sludge. Oxygen transfer changes dynamically in activated sludge tanks depending on wastewater influent and process characteristics and this variance can result in failures of process design and operation (Amaral et al., 2017). Thus, modelling oxygen transfer parameters such as the α-factor could provide a useful tool to improve design and operation of aeration systems. Currently, no generally applicable oxygen transfer model exists (Baquero-Rodríguez et al., 2018). Previous approaches are limited to a small number of parameters describing the process, some of which are difficult to reliably quantify (e.g., sludge retention time) or measure in practice (e.g., bubble size). In recent years, data-driven methods were utilized in several modelling applications in the field of wastewater treatment (Alejo, 2021; Tyralis et al., 2019). In this research work, three different WWTPs with four different AS stages were examined in long-term ex situ off-gas testing which provided a comprehensive dataset suitable for the application of data-driven methods. The main idea behind P3 was to model the α-factor with a supervised machine learning algorithm based on operating data typically available for WWTP operators. P3 discusses the modelling process to predict the α-factor and limitations of this methodology.

Prior to these papers, this thesis summarizes the state of the art in the field of aeration technology in chapter 2. The chapter presents concepts and terminology regarding the measurement and modelling of oxygen transfer parameters which is the main subject of this research work. Based on this background the objective of each paper is described in chapter 3. The chapter also outlines how the individual papers are linked with each other. Chapter 4 contains the papers that form the cumulative part of this thesis. Finally, the conclusions in chapter 5 cover the contributions of each individual paper in the context of this research work and combined findings derived from the papers. The chapter is concluded by an outlook proposing prospective future work beyond this thesis.

2 State of the Art – Aeration Technology

Treatment of wastewater in municipal wastewater treatment plants (WWTP) uses mechanical and biological processes. Biological wastewater treatment is typically performed in the activated sludge process that has been utilized for more than 100 years (Jenkins and Wanner, 2014). Microorganisms are grown in an activated sludge tank using wastewater contaminants in the influent as a substrate. Water and sludge flows are controlled to maintain this microbial community at a high concentration to facilitate an effective removal of substrate via biological removal or solids separation in the subsequent clarifier. This solids separation returns activated sludge into the activated sludge tank or, partially, removes waste activated sludge (WAS) from the process to maintain a stable concentration of activated sludge in the tank measured as total suspended solids (TSS). Supernatant from the clarifier is either directed to further treatment stages or discharged from the WWTP.

The activated sludge process can be separated into anaerobic and aerobic stages. The oxygen supply to microorganisms for aerobic biological treatment is typically provided by submerged diffusers or a mechanical surface aeration system that generate gas-liquid interfaces for oxygen transfer. For submerged aeration systems ambient air is compressed by blowers and delivered to diffusers installed on the bottom of an activated sludge tank where air bubbles are released through small orifices. Oxygen is transferred from the gas phase to the soluble phase as the bubbles rise in the activated sludge. Mechanical aeration systems submerge air below the surface of the activated sludge and shear the liquid surface into droplets with mixing devices to increase gas-liquid interface. Nowadays, fine-bubble diffusers are the most common aeration technology as they can be operated at higher standard aeration efficiency (SAE) which describes oxygen transfer per unit energy consumed (Wagner and Stenstrom, 2014). Diffusers are typically installed to evenly cover the bottom of the aerobic stage of an activated sludge tank. Here, the oxygen transfer is a three-phase system with air as the gas phase, sludge as the solid phase, and wastewater as the soluble phase. Aeration systems must provide oxygen for aerobic treatment and sufficient mixing to prevent sedimentation of activated sludge in the tank. Anoxic tank zones therefore typically require additional mixing devices. Aeration is an energy-intensive process, even though numerous advances in diffuser design and operation of WWTPs have been made. In most wastewater treatment plants (WWTP) it accounts for more than half of the net energy consumption (Baquero-Rodríguez et al., 2018; Reardon, 1995; Rosso et al., 2011). Energy costs for wastewater treatment account for approximately 0.7 to 1 % of total energy consumption in Germany (Maktabifard et al., 2018).

In general, two aspects are optimized for energy-efficient operation of submerged aeration systems. Firstly, the oxygen transfer efficiency (OTE) should be as high as possible, i.e., the amount of oxygen transferred at bubble rise from release at the diffuser to the activated sludge tank surface. OTE is usually higher at smaller bubble diameters that feature a larger specific interface between gas and liquid phase produce a low flow regime with less turbulence than coarse bubbles. Secondly, the diffuser pressure loss should be as low as possible, i.e., the counter-pressure applied by the perforated diffuser membranes. Diffuser pressure loss is typically higher for diffuser membranes with smaller slits producing finer bubbles. Therefore, the aim of diffuser design is to optimize the opposing parameters OTE and diffuser pressure loss. Definition and measurement of these parameters is described in chapter 2.1. In addition to diffuser specific parameters, the design and operation of the activated sludge tank can have a significant effect on OTE as well as described in chapter 2.2.

2.1 Quantifying Oxygen Transfer

Standard testing procedures in technical guidelines describe how to determine the oxygen transfer of an aeration system. The procedures aim to reduce the overall uncertainty to the instrument's errors and thereby increase comparability of results from different testing facilities. There are methods to measure oxygen transfer in clean water and activated sludge. Table 1 lists available technical guidelines for oxygen transfer testing and where they are applicable.

Table 1: Overview of technical guidelines for oxygen transfer testing

Region	Clean Water	Activated Sludge
European Norm	EN 12255-15 (2003)	-
German Standard	DWA-M 209 (2007)	DWA-M 209 (2007)
American Standard	ASCE 2-06 (2006)	ASCE 18-18 (2018)

The quantification of oxygen transfer parameters is crucial for proper design and operation of aeration systems which is further described in chapter Modelling Oxygen Transfer Dynamics. The methods in technical guidelines describe procedures to empirically determine the oxygen mass transfer from gas to liquid. Several mass transfer models exist in the theory of gas-liquid absorption with the stagnant two-film model (Lewis and Whitman, 1924), the penetration theory (Higbie, 1935) and the surface renewal model (Danckwerts, 1951) being the most common. The mass transfer models used in technical guidelines listed in Table 1 assume that interfacial films are stagnant with only diffusional transport across the interface (Lewis and Whitman, 1924) as described in the following chapters. It is worth noting, however, that more advanced gas transfer theories consider non-stagnant gas-liquid interfaces. Higbie's penetration theory suggests a continuous regeneration of the gas-liquid surface describing an exchange of fluid at the bubble's interface. Similarly, Danckwert's surface renewal model refines the theory by also considering the age of the gas-liquid surface. Applying the theories requires the consideration of parameters such as the velocity of adsorption and interfacial gas-liquid, the surface element contact time, and the bubble diameter. While this is beneficial to account for the effect of turbulence in a tank on oxygen transfer, its shortcoming is that these parameters can generally not be measured in full-scale setups. Consequently, the intricate details of non-stagnant gas-liquid interfaces and thereof resulting bubble mechanics are not included in oxygen transfer parameters calculated according to technical guidelines. In practice, this simplification can lead to a bias of results, e.g., when performing clean water and activated sludge comparison tests under different hydraulic conditions in the same test tank.

2.1.1 Oxygen Transfer Testing in Clean Water

Oxygen transfer testing of aeration systems in clean water is a standardized procedure to quantify the performance of diffusers. The procedure is conducted in a two-phase system with air bubbles rising in tap water. This provides the best reproducibility of test results because there are fewer variations influencing the oxygen transfer in clean water than in activated sludge. The test method determines the mass transfer coefficient k_La and the oxygen saturation concentration C^*_∞ which are the basis for all further oxygen transfer parameters. The procedure can be conducted either as a desorption or absorption test. For desorption testing, dissolved oxygen concentration is initially increased with pure oxygen and afterwards stripped with air until the oxygen saturation concentration is reached. For absorption testing, dissolved oxygen is initially chemically absorbed to lower DO close to zero and afterwards reoxygenated until the oxygen saturation concentration is reached. This results in a typical DO profile during an oxygen transfer test that follows a saturation curve, as described by the differential form:

$$\frac{dC}{dt} = k_L a \cdot (C^*_\infty - \mathrm{C_t}) \tag{1}$$

which yields the following exponential equation:

$$C_\mathrm{t} = C^*_\infty \text{ - } (C^*_\infty \text{- } C_0) \cdot e^{-k_L a \cdot (t - t_0)} \tag{2}$$

with:

k_La:	apparent volumetric mass transfer coefficient under test conditions (h^{-1})
C_t:	dissolved oxygen concentration at time t ($mg \cdot L^{-1}$)
C^*_∞:	steady-state dissolved oxygen saturation concentration as time approaches infinity ($mg \cdot L^{-1}$)
C_0:	dissolved oxygen concentration at start of test (t = 0) ($mg \cdot L^{-1}$)

Figure 1 shows the DO profile of a desorption oxygen transfer test. The collected data is iteratively fitted by a nonlinear least squares (NLS) regression function of the form of equation 2 (blue line). This curve converges with the oxygen saturation concentration C^*_∞ (blue dotted line) under test conditions. Residuals represent the error of the NLS regression as the difference between the fitted value and each measured value at the same duration timestamp. Evenly and randomly distributed residuals indicate a reliably conducted test under sufficient mixing conditions.

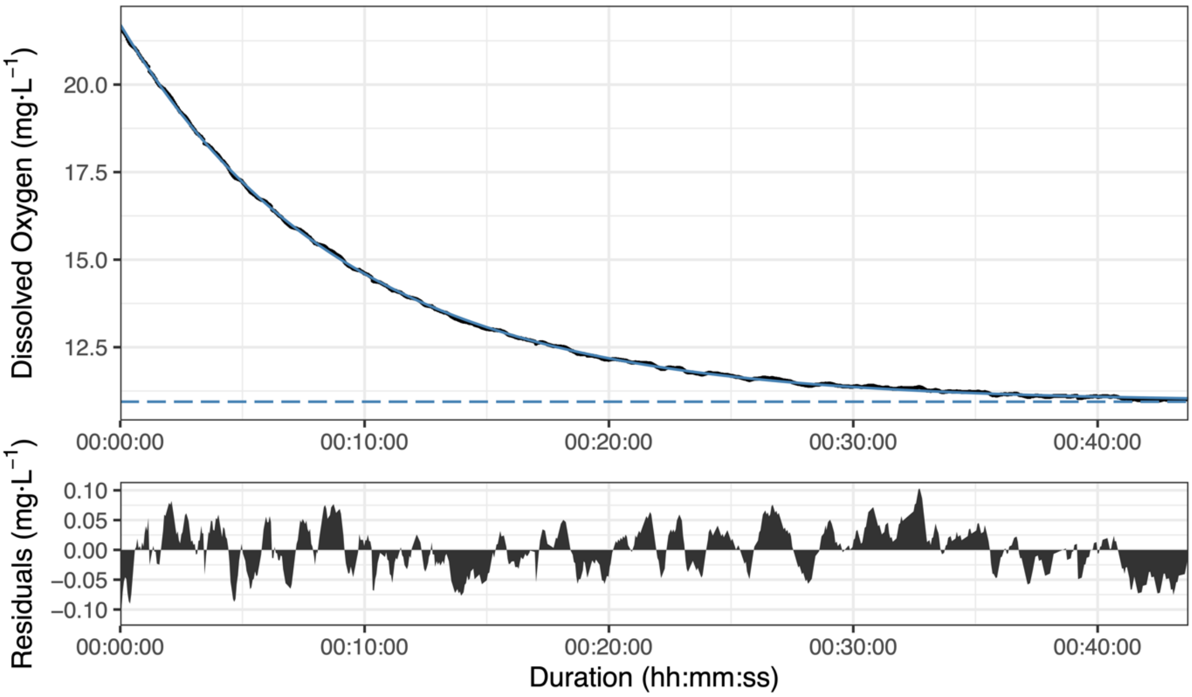

Figure 1: Dissolved oxygen profile of a desorption oxygen transfer test

Water temperature, atmospheric pressure, and salinity of test water influence oxygen transfer and are therefore standardized. Temperature and salinity of volumetric mass transfer coefficient k_La are corrected to 20 °C in k_La_{20} and 1,000 mg·L^{-1} in $k_La_{20,1000}$:

$$k_La_{20} = k_La \cdot \theta^{(20-T)} \tag{3}$$

$$k_La_{20,1000} = \frac{1.1 \cdot k_La_{20}}{1 + 0.1 \cdot \frac{TDS}{1{,}000}} \tag{4}$$

with:

θ: Temperature correction factor (theta), θ=1.024 (-)

TDS: Total dissolved solids, often approximated with the electrical conductivity (EC); a common conversion for TDS in mg·L^{-1} is:
EC (in µS·cm^{-1}) · 2,000 mg·L^{-1} TDS / 3,000 µS·cm^{-1} (DWA, 2007)

k_La_{20}: volumetric mass transfer coefficient at 20 °C (h^{-1})

$k_La_{20,1000}$: volumetric mass transfer coefficient at 20 °C and 1,000 mg·L^{-1} TDS (h^{-1})

Water temperature and atmospheric pressure of oxygen saturation concentration C^*_∞ are corrected to 20 °C and 101.325 kPa in C^*_{20}:

$$C_{S,T,St} = \frac{2234.34\ (\mathrm{mg \cdot L^{-1} \cdot °}C)}{(T_w + 45.93)^{1.31403}} \tag{5}$$

$$C^*_{20} = C^*_\infty \cdot \tau \cdot \Omega = C_{S,md} \cdot \frac{C_{S,T,St}}{9.09} \cdot \frac{p_{atm}}{101.325} \tag{6}$$

with:

T_w: Water temperature (°C)

$C_{S,T,St}$: Oxygen saturation concentration at water temperature Tw (mg·L^{-1})

$C_{S,md}$: Oxygen saturation concentration at mid-depth of tank 1(mg·L^{-1})

τ: Temperature correction (tau) of effective saturation concentration (-)

Ω: Pressure correction (omega) of effective saturation concentration (-)

p_{atm}: Atmospheric pressure (kPa)

C^*_{20}: Standardized effective oxygen saturation at process conditions (mg·L^{-1})

ASCE 18-18 refers to tabulated values by Benson and Krause Jr (1984), the polynomial above is defined in DWA-M 229-1 (2017)and calculates these values. Typical oxygen transfer parameters in clean water (cw) are the standard oxygen transfer rate ($SOTR_{cw}$) and standard oxygen transfer efficiency ($SOTE_{cw}$):

$$SOTR_{cw} = \frac{\mathrm{V \cdot k_La_{20,1000}} \cdot C^*_{20}}{1{,}000\ (\mathrm{g \cdot kg^{-1}})} \tag{7}$$

$$SOTE_{cw} = \frac{100\ \% \ \cdot SOTR_{cw}}{Q_{AFR} \cdot 0.299\ (\mathrm{kg} \cdot m^3)} \tag{7}$$

with:

V: Volume of test tank (m^3)

Q_{AFR}: Airflow rate, usually at standard temperature and pressure (Nm^3·h^{-1})

$SOTR_{cw}$: Standard oxygen transfer rate (kg·h^{-1})

$SOTE_{cw}$: Standard oxygen transfer efficiency (%)

0.299: Mass of oxygen per volume of ambient air (1.429 kg O_2 per m^3 in pure oxygen equals 0.299 kg O_2·m^3 at 20.946 % oxygen concentration in ambient air)

It is worth noting that this testing procedure quantifies the overall oxygen transfer occurring in a test tank. The analysis of a DO profile in this non-steady state testing yields a volumetric mass transfer coefficient k_La. In this parameter k_L corresponds to the gas-liquid diffusion coefficient and a corresponds to the specific surface area of the rising air bubbles (surface area per volume). Measuring diffusion at this phase boundary layer or measuring its surface area is usually not possible in practice. Therefore, the volumetric mass transfer coefficient k_La is used as a reference value to characterize the oxygen transfer (Eckenfelder Jr, 1959).

2.1.2 Off-gas testing in Activated Sludge

Oxygen transfer in full-scale activated sludge tanks can be determined with non-steady state methods like the clean water testing procedure described before. An absorption or desorption DO profile is created by changing power levels of blowers or adding H_2O_2, similarly to the pure oxygen desorption method in clean water testing. These methods, however, rely on constant oxygen uptake and the ongoing biodegradation of wastewater contaminants, the uncertain measurement of oxygen uptake rate, and the continuous wastewater inflow can introduce additional errors.

Another established method is the off-gas measurement. In addition to dissolved oxygen sensors, it requires a gas analyzer to record oxygen concentrations in the off-gas of activated sludge tanks. With the in situ method off-gas is collected from fixed or floating hoods on the top of the activated sludge tank surface. An ex situ alternative exists where activated sludge is transferred into an adjacent aeration column. Both off-gas methods use a mass balance to calculate oxygen transfer efficiency under process conditions OTE_f as follows:

$$OTE_f = \frac{MR_i - \left(\frac{O_{2,e}}{1 - O_{2,e} - CO_{2,e}}\right)}{MR_i} \tag{8}$$

with:

$O_{2,e}$: Oxygen concentration in off-gas, without water vapor and particle-free (%)

$CO_{2,e}$: Carbon dioxide concentration in off-gas, without water vapor and particle-free (%)

MR_i: Molar ratio of oxygen to inert substances (-)

OTE_f: Oxygen transfer efficiency under process conditions (%)

$$MR_i = \frac{\frac{O_{2,in}}{1 - CO_{2,in}}}{1 - \frac{O_{2,in}}{1 - CO_{2,in}}} = 0.265 \tag{9}$$

with:

$O_{2,in}$: Inlet oxygen concentration of 20.946 %

$CO_{2,in}$: Inlet carbon dioxide concentration of 0.0407 %

The parameter is calculated depending on the gas analyzer output and/or off-gas conditioning. In the presented calculation off-gas concentrations were already pressure corrected to atmospheric pressure. For other variants the reader is referred to ASCE 18-18 (2018) or DWA-M 209 (2007). For better comparison between measurements the oxygen transfer efficiency under process conditions is standardized to a water temperature of 20 °C, atmospheric pressure of 101.325 kPa, and zero DO to $SOTE_{pw}$ according to the following equations:

$$OTE_{sp,20} = \frac{OTE_f}{C_{20}^* - C_t} \cdot \theta^{20-T_w} \tag{10}$$

with:

C_t: Dissolved oxygen concentration in the ex situ column (mg·L^{-1})

C^*_{20}: Standardized effective oxygen saturation at process conditions (mg·L^{-1})

θ: Temperature correction factor (theta); θ = 1.024 (-)

T_w: Water temperature (°C)

$OTE_{sp,20}$: Oxygen transfer efficiency per unit of driving force at std. conditions (%/(mg·L^{-1}))

The β-factor is the ratio of oxygen saturation in process water to clean water equivalent conditions of water temperature partial pressure:

$$\beta = 1.00 - 0.01 \cdot \frac{TDS}{1{,}000\ \mathrm{mg} \cdot \mathrm{L}^{-1}} \tag{11}$$

with:

TDS: Total dissolved solids (mg·L^{-1})

β: β-factor (beta) (-)

Combining these parameters yields the standardized oxygen transfer efficiency under process conditions:

$$SOTE_{pw} = OTE_{sp20} \cdot C_{20}^* \cdot \beta \tag{12}$$

with:

C^*_{20}: Standardized effective oxygen saturation at process conditions (mg·L^{-1})

$SOTE_{pw}$: Standard oxygen transfer efficiency under process conditions (%)

Oxygen transfer under process conditions in activated sludge is inhibited by several influencing factors as described in chapter *Influences on Oxygen Transfer in the Activated Sludge Process*. The sum of these influences is combined in the α-factor and calculated as a simple ratio of oxygen transfer efficiency in process water and clean water:

$$\alpha = \frac{SOTE_{pw}}{SOTE_{cw}} \tag{13}$$

with:

$SOTE_{cw}$: Standard oxygen transfer efficiency in clean water (%)

α: α-factor (-)

Diffuser related factors, such as fouling, scaling, and aging that reduce oxygen transfer are often separately considered by the fouling factor F and therefore stated as αF-factor, see section 2.2.6. Because wastewater contaminants inhibiting oxygen transfer cannot be determined a priori the resultant α-factor during operation may only be determined a posteriori with the above equations. Chapter 2.2 outlines these influences on oxygen transfer in the activated sludge process.

2.2 Influences on Oxygen Transfer in the Activated Sludge Process

Oxygen transfer in activated sludge is a highly dynamic process. Its variation affects air requirements of the activated sludge tank and thus operational cost. Influences on minimum and peak air requirement also affect equipment sizing and the design of aeration systems. The gas-liquid mass transfer in activated sludge tanks is measured as an oxygen transfer rate or efficiency (SOTR or SOTE) or, when compared with oxygen transfer in clean water, as an α-factor. Earliest studies were conducted several decades ago by Kessener and Ribbius (1934) and Eckenfelder and Barnhart (1961). Since then, numerous investigators have studied the influences on oxygen transfer in the activated sludge process. Modelling the mass transfer is particularly challenging in the three-phase system as many factors are difficult to measure. This chapter gives an overview about influences on oxygen transfer separated into sections for solid, liquid, and gas phase. In addition, the effect of standardization parameters, activated sludge process characteristics related to operation, and diffuser related characteristics are summarized. It is worth noting that many parameters are interrelated with each other and could therefore be included in multiple sections.

2.2.1 Solid Phase

The solid phase in activated sludge comprises biomass in the form of activated sludge flocs and particulate matter not removed from wastewater inflow in mechanical wastewater treatment steps. This solid matter can be categorized into a wide range of sizes from colloidal (0.001 to 1 µm), supracolloidal (1 to 100 µm), to settleable contaminants (> 100 µm) (Levine et al., 1991). In activated sludge it is typically measured as total suspended solids (TSS), also known as mixed liquor suspended solids (MLSS) with filters with 0.45 µm pore width. The presence of a solid phase distinguishes oxygen transfer in the activated sludge from clean water conditions. Hence, its effect on oxygen transfer efficiency in aeration tanks has been examined extensively in the past.

Monitoring in Activated Sludge Tanks

WWTP operators require information about the total biomass concentration within an activated sludge tank to monitor and control the treatment process. In addition, settleability of the activated sludge is monitored to ensure a proper phase separation in clarifiers.

Total suspended solids (TSS in $g \cdot L^{-1}$) and total solids (TS in $g \cdot L^{-1}$) are typically measured in regular laboratory analysis according to the standard method that is valid in the location of the WWTP. In principle, TS is determined by the remaining weight of a sample after evaporation in an oven. For TSS the sample is filtered through a glass-fiber filter before the residue is dried to constant weight. Total dissolved solids (TDS) are determined when the filtrate is examined by the same principle. TS is therefore the sum of TSS and TDS. Principles are described in standards such as Standard Methods for the Examination of Water and Wastewater (AWWA, 2017), EN 12880 (2000), and EN 15935 (2021). Online sensors can record data with a higher temporal resolution than laboratory-based analysis. Turbidity sensors measure the reduction of transparency in a liquid caused by the presence of undissolved matter as described in ISO 7027-1 (2016). Sensors can be calibrated for the use in activated sludge with different ranges of TSS concentration depending on the type of sludge.

Settleability of activated sludge is characterized by sludge volume (in $mL \cdot L^{-1}$) and sludge volume index (SVI in $mL \cdot g^{-1}$). The sludge volume is the remaining volume of an activated sludge sample after 30 minutes settlement. SVI relates sludge volume to total suspended solids to describe the settleability of an activated sludge sample. The parameter is often determined manually although automated setups exist. Data is recorded with a low temporal resolution due to the settling time of 30 minutes.

All laboratory methods face the challenge to take a representative sample of an activated sludge tank. Samples of the activated sludge tank are often taken adjacent to position of online sensors for validation and calibration purposes. Likewise, an online sensor is a point measurement within a three-dimensional reactor. Consequently, depending on the type of reactor and the mixing conditions the position of an online sensor or a sampling location might not represent the whole activated sludge tank. Rieger (2012) list potential sources of errors during sampling and of online sensors. These sources of errors are site-specific and can increase the measurement uncertainty of an analysis above the expected level listed in technical standards.

Interacting Mechanisms influencing Oxygen Transfer

It is often stated that TSS is inversely correlated with the α-factor. This relationship has been demonstrated extensively for membrane bioreactors (MBR) where sludge rheology at TSS up to 30 $g \cdot L^{-1}$ is the primary influence on oxygen transfer (Cornel et al., 2003; Germain et al., 2007; Krampe and Krauth, 2003). Henkel (2010) adds that especially the organic fraction of suspended solids (mixed liquor volatile suspended solids - MLVSS) causes oxygen transfer inhibition. Based on these studies the inverse relationship measured in MBRs was extrapolated to describe mechanisms in CAS systems, where typical TSS concentrations are lower than 6 $g \cdot L^{-1}$. More recent studies additionally consider the concept of biosorption that describes interactions of soluble and solid substances. This link between the solid and liquid phase is the adsorption of oxygen transfer inhibiting soluble substances on the suspended sludge flocs. Biosorption therefore decreases concentration of organic substances in the liquid phase and hence also reduces accumulation of oxygen transfer inhibiting substances at the gas-liquid interface (Ahmed et al., 2021a; Odize, 2018). Under conditions where biosorption is the dominant impact on oxygen transfer, higher TSS can be positively correlated with the α-factor as biosorption is increasing. Baquero-Rodríguez et al. (2018) formulate this positive correlation between TSS concentrations up to 6 $g \cdot L^{-1}$ and the α-factor. However, Ahmed et al. (2021b) conclude that so far no robust relationship between TSS and the α-factor exists for CAS systems.

Other parameters to describe characteristics of the solid phase exist and have been related to oxygen transfer. These parameters outlined below have in common that they are not typically measured in full-scale WWTP operation and therefore remain unknown to the operator.

- Solid holdup of a three-phase system is a common parameter in chemical engineering and describes the bulk volume of solids as a percentage of the total volume. Sludge volume does not represent solid holdup because particle mass is not densely separated. Therefore, Henkel et al. (2011) introduced hydrostatic floc volume (HFV) as an alternative where biomass is inactivated with a biocide to prevent denitrification and settling time is increased to 48 hours. Like TSS, an increase of HFV reduced the α-factor in Henkel's experiments in MBRs.
- TSS does not distinguish organic from inorganic matter. Henkel (2010) has demonstrated that the organic fraction VSS (volatile suspended solids) primarily inhibits oxygen transfer. However, this organic matter is not divided into fractions of active and inactive biomass. These aspects directly affect the oxygen uptake rate as a higher proportion of active biomass also increases total oxygen consumption

under otherwise constant conditions. The resultant difference of DO also changes the dissolved oxygen diffusion gradient and thus indirectly affects oxygen transfer efficiency.

- Extracellular polymeric substances (EPS) are an important component for floc structure. EPS are a mixture of polymers excreted by microorganisms, adsorbed organic matter from wastewater, and substances produced from cell lysis that hold microbial aggregates together in a three-dimensional matrix (Sheng et al., 2010). Steinmetz (1996) demonstrated a negative correlation between EPS concentration and the α-factor in batch-scale experiments. Germain et al. (2007) conducted pilot and full-scale experiments with MBRs and in addition to EPS concentration considered the effect of chemical oxygen demand (COD) concentration of the soluble microbial products, and TSS, on the α-factor. They found that in MBRs TSS concentration still was the dominating influence on α-factor decrease, followed by COD concentration of the soluble microbial products and EPS concentration.
- TSS does not distinguish particle size distribution of flocs in activated sludge. The link between carbon removal and particle size distribution has been examined before in wastewater applications (Arslan-Alaton et al., 2009; Houghton et al., 2002; Levine et al., 1991). Li and Stenstrom (2017) examined particle size distribution of five full-scale WWTPs and found a positive correlation with SRT as mean particle size increased with higher SRT. As higher SRT generally also increases oxygen transfer efficiency, particle size distribution can be indirectly related to oxygen transfer. However, implications of particle size distribution on oxygen transfer have not been examined so far.
- Finally, it is worth noting that the dosage of chemicals for flocculation and precipitation impact floc structure, adsorption capacity, settleability, and total suspended solids concentration (Rahman et al., 2016; Schuler et al., 2001). So far, no comprehensive research has been conducted to examine their influence on oxygen transfer.

It is worth emphasizing that the typical monitoring of the solid phase in an activated sludge tank is missing to describe several of its characteristics. The mechanisms depicted above alter the liquid-solid interface, thus also influencing the gas-liquid and gas-solid interface. As a conclusion, oxygen transfer models based exclusively on TSS as the typically available information about the solid phase in an activated sludge tank greatly oversimplify the three-phase system.

2.2.2 Liquid Phase

The liquid phase in activated sludge can be categorized as bulk water and as water held within capillaries or on particle surfaces by adsorption (Katsiris and Kouzeli-Katsiri, 1987). In activated sludge systems the liquid holdup has the largest share of total volume compared to solid and gas holdup. Besides water, the liquid phase consists of soluble substances such as dissolved salts and non-dissolved matter like colloids and free moving bacteria. Among the most important oxygen transfer inhibitors in the liquid phase are soluble fractions of COD in general and surfactants in specific.

Monitoring in Activated Sludge Tanks

Most soluble contaminants are typically characterized in the activated sludge tank's influent and effluent flows with online sensors and/or routinely performed laboratory analytics. Monitoring parameters representing carbon, nitrogen, and phosphorus concentrations is required to control the activated sludge process and evaluate its treatment performance for regulatory compliance of emissions. Among the most common parameters are total organic carbon (TOC), chemical oxygen demand (COD), and biological oxygen demand (BOD) to account for carbon as well as total Kjeldahl nitrogen (TKN), ammonia NH_4-N, nitrate NO_3-N, and total nitrogen (N_{tot}) for nitrogen and phosphate (PO_4-P), and total phosphorus (P_{tot}) for phosphorus. These parameters are defined in basic literature (inter alia, Henze et al., 2008; Tchobanoglous et al., 2014) and analysis procedures are described in technical standards such as Standard Methods for the Examination of Water and Wastewater (AWWA, 2017). Vanrolleghem and Lee (2003) give an overview of available instruments for online measurement. The scope of analysis at a WWTP to monitor the process varies with plant size, its process requirements, and local regulatory requirements. Rieger (2012) outlines sources of errors due to installation of instruments for online measurement and sampling procedures and describes methods for data reconciliation for modelling applications.

However, unlike the solid phase that is routinely measured as TSS in activated sludge tanks, it is not common to examine soluble wastewater contaminants in the liquid phase of the activated sludge. For the aeration process this means that concentrations of oxygen transfer inhibiting soluble substances in the three-phase system are largely unknown at the point of impact, i.e., the bubble rising in activated sludge. Instead, current modelling of oxygen transfer requires assumptions about dilution, distribution, and biodegradation of oxygen transfer inhibiting contaminants within the activated sludge tank. These are based on the available information about influent and effluent parameters as well as activated sludge process characteristics, see section 2.3.

Aeration control systems require online sensors for process control. Electrochemical cells and optical sensors can monitor DO in aerobic tank zones. To control aeration for nutrient removal, in-situ ion-selective electrodes (ISE) can monitor ammonium and nitrate. These are an alternative to ex situ automated wet chemistry analyzers in influent and effluent with a faster response time (Vanrolleghem and Lee, 2003). The location of these sensors in the activated sludge tank or its influent and effluent flows depends of the aeration control scheme (Åmand et al., 2013).

COD Wastewater Load

Overall, a negative correlation between chemical oxygen demand (COD) load in wastewater inflow and oxygen transfer efficiency in the activated sludge tank exists. This has been shown by Günkel-Lange (2013) for four different WWTP treatment processes in pilot-scale, by Leu et al. (2009) based on real-time off-gas measurements in a full-scale CAS system, and by Germain et al. (2007) for pilot and full scale MBRs. Jiang et al. (2017) concluded a negative logarithmic relationship between the α-factor and COD based on additional data from full-scale treatment plants with higher COD load. Ahmed et al. (2021a) further distinguished COD load into acetate as readily biodegradable substrate and cellulose as slowly biodegradable substrate in batch-scale experiments. They found that, depending on airflow rate, readily biodegradable acetate decreased α-factor more than slowly biodegradable cellulose. The drastic inhibition of oxygen transfer by surfactants as a fraction of overall COD is outlined separately in the next section.

In contrast to COD influent load, Steinmetz (1996) found no correlation between dissolved organic carbon (DOC) in the activated sludge and the α-factor. However, she concluded that DOC samples did not include wastewater contaminants adsorbed to sludge flocs due to the filtration process. Nonetheless, these findings emphasize that considering the interaction of oxygen transfer inhibiting soluble substances and the solid phase is crucial to understand the overall oxygen transfer process, see section 2.2.1.

The Role of Surfactants

The oxygen transfer inhibiting effect of surfactants has long been observed (Kessener and Ribbius, 1934; Mancy and Okun, 1960) and therefore has been the subject of numerous studies in aeration technology (inter alia: Henkel, 2010; Jimenez et al., 2014; Loubière and Hébrard, 2004; Rosso, 2005; Rosso and Stenstrom, 2006; Sardeing et al., 2006; Wagner and Pöpel, 1996). Surfactants such as fatty acids, soaps, and detergents are amphiphilic substances with hydrophilic and lipophilic properties, which have two effects on gas-liquid interfaces. On the one hand, a decrease of dynamic surface tension produces smaller bubbles, thus increasing specific interfacial area and oxygen transfer. On the other hand, the surfactants accumulate on the hydrophobic gas-liquid interface and reduce diffusion k_L and renewal of interfacial area as bubbles become more rigid (Eckenfelder Jr, 1959; Mancy and Okun, 1960). The latter effect inhibiting oxygen transfer is usually predominant for fine-bubble diffusers with lower flow regimes and rise velocities of bubbles. Residence times of fine bubbles are longer, and surfactants can accumulate more on the gas-liquid interface than under more turbulent conditions produced by coarse bubble diffusers or high airflow rates (Rosso and Stenstrom, 2006a; Stenstrom and Gilbert, 1981). Although, surfactants usually account for a small fraction of overall COD in wastewater inflow, their effect on oxygen transfer inhibition can be drastic. Consequently, modifications of the activated sludge process design that reduce the negative effect of surfactants on oxygen transfer have been investigated, see section 2.2.5.

2.2.3 Gas Phase

Even though aeration systems provide the gas phase in the three-phase system of aerobic activated sludge treatment, not much is known about the gas holdup's characteristics in full-scale applications.

Gas Holdup in a three-phase System

The gas holdup is defined as a percentage share of gas phase volume of total volume of the three-phase system. Babaei et al. (2015) used electrical resistance tomography to measure gas holdup in three-phase systems with various total suspended solids concentrations (TSS between 0.712 – 15.86 $g \cdot L^{-1}$) and various superficial gas velocities (airflow rate between 0.163 – 1.303 $cm \cdot s^{-1}$) in a bubble column. As expected, airflow rate and gas holdup followed a linear trend. However, for increasing TSS the overall gas holdup increased until about 2.15 $g \cdot L^{-1}$ and was followed by a decreasing gas holdup for higher TSS concentrations. This trend was primarily attributed to the effect of increasing solid holdup on the rheological characteristics of the activated sludge. Transferring findings by Babaei et al. (2015) allows to estimate the overall gas holdup of typical activated sludge processes as well below 1 %, even at high-rate activated sludge systems with volume specific airflow rates of about 2 $Nm^3 \cdot m^{-3} \cdot h^{-1}$. Consequently, in a CAS aeration tank operated at about 0.5 $Nm^3 \cdot m^{-3} \cdot h^{-1}$ the overall gas holdup would be four times lower.

Bubble Characteristics

Any additional characteristics of the gas holdup such as bubble shape, size and rise velocity, or coalescence of bubbles is impossible to measure in a three-phase system. Some relationships between these characteristics and other measurable quantities were studied before. Typically, for a given orifice size the higher the airflow rate the larger the bubble diameter (Clift et al., 1978). Additionally, perforated slits of fine-bubble diffuser membranes widen at higher airflow rates, thus releasing larger bubbles (U.S. EPA, 1989). Similarly, the higher the airflow rate the higher the overall turbulence due to the bubble flow regime will be in an activated sludge tank. In general oxygen transfer efficiency of coarse bubble aeration is lower than fine bubble aeration (Groves et al., 1992). But because turbulence influences renewal of phase boundary, this becomes increasingly important at high concentrations of surfactants that inhibit gas transfer at the gas-liquid interface. In this case, higher turbulence produced by coarse bubbles can improve oxygen transfer (Rosso, 2005). These examples have in common that they are related to diffuser specifications and are further outlined in section 2.2.6.

Monitoring in Activated Sludge Tanks

Besides these diffuser related characteristics, measurement of the airflow rate is the only parameter that characterizes the gas phase in the activated sludge process. Instruments for flow measurement can record airflow rate with low response time. An interval of 1 to 5 minutes is required for an optimized aeration control (Rieger, 2012), so temporal resolution of data is not limited to monitor the process. However, many aeration control schemes in practice do not measure the spatial distribution of total airflow rate within an activated sludge tank. Instead, only a total airflow rate for a treatment stage is recorded without individual airflows into different aeration zones. For power-minimizing control strategies, pressure in air headers is monitored in each aeration zone. For example, when a most-open-valve (MOV) principle is used, valves for each aeration zone are controlled to minimize pressure in air headers by almost completely opening the most open valve (Alex et al., 2002). Nonetheless, gas holdup or airflow rate cannot be estimated based on pressure in air headers for each aeration zone. In addition, most other aeration control strategies focus on monitoring oxygen in the dissolved state instead of the airflow rate (Olsson et al., 2018). In any case, state-of-the-art monitoring and control of aeration systems in activated sludge processes does not record characteristics of gas phase. Consequently, including information about the gas phase to model oxygen transfer processes is currently limited as described in chapter 2.3.

2.2.4 Standardization Parameters

Oxygen transfer parameters are standardized to a water temperature of 20 °C and atmospheric pressure of 101.325 kPa (ASCE 2-06, 2007; DWA-M 209, 2007; EN 12255-15, 2003). German technical guideline DWA-M 209 (2007) also standardizes total dissolved solids to 1,000 $mg \cdot L^{-1}$ and approximates the salinity by electric conductivity. Operating data for standardization is recorded at most WWTPs. The effect on oxygen transfer is summarized below.

Water Temperature

Higher water temperature in activated sludge not only affects oxygen uptake of microorganisms due to increased activity but also decreases solubility of oxygen. The effect of water temperature on oxygen saturation concentration is standardized to 20 °C in oxygen transfer parameters using a temperature correction factor θ (theta) = 1.024. This parameter was empirically determined in a range between 1.008 to 1.047 (Stenstrom and Gilbert, 1981), with ranges from 1.020 to 1.028 deemed reasonable according to United States Environmental Protection Agency (1989). It is worth noting that an increase of water temperature often correlates with ambient air temperature which also decreases the capacities of blowers (Jenkins, 2013).

Atmospheric Pressure

Atmospheric pressure directly affects oxygen saturation concentration and is therefore standardized to 101.325 kPa in a linear fashion to determine standard oxygen transfer parameters. Its impact on the oxygen transfer efficiency has not been studied yet (Baquero-Rodríguez et al., 2018). The parameter is important to evaluate blower performance and resultant energy requirements as air density and atmospheric pressure are correlated. This evaluation depends on blower type, control techniques, and operating conditions (Rosso, 2018).

Salinity

Salinity as total dissolved solids (TDS) can be approximated by electrical conductivity. DWA-M 209 (2007) applies a conversion from TDS to electrical conductivity of $2\ mg \cdot L^{-1}$ TDS = $3\ \mu S \cdot cm^{-1}$. This factor of 0.67 was confirmed by Behnisch et al. (2021) who determined a mean of 0.7 for different salts, whereas AWWA standard methods list a range between 0.55 and 0.9 (AWWA, 2017). Higher salinity slightly decreases oxygen transfer saturation (Benson and Krause Jr, 1984). This effect is negligible and therefore not considered in clean water and off-gas testing. On the other hand, the generally positive effect of salts on oxygen transfer due to inhibition of bubble coalescence is considered when standardizing k_La_{20} to a TDS concentration of $1{,}000\ mg \cdot L^{-1}$ for $k_La_{20,1000}$ in clean water. Nonetheless, the effect on α-factor determination under process conditions is negligible at the salt concentrations expected in municipal wastewater. Some uncertainty about this standardization with the β-factor remains, as Eckenfelder et al. (1956) report β-factors as approximately 0.95 in municipal wastewater and ASCE 18-18 states variations between 0.8 to 1.0 are possible, but that β-factor is generally close to 1.0. Baquero-Rodríguez et al. (2018) conclude that the β-factor for wastewater with a TDS concentration below $1{,}500\ mg \cdot L^{-1}$ is approximately 0.99 and for industrial wastewater with about $10{,}000\ mg \cdot L^{-1}$ is estimated at 0.94.

Relevance of Standardization Parameters for Oxygen Transfer Models

Discussing standardization parameters in the context of oxygen transfer modelling is relevant for two reasons. Firstly, the parameters are determined empirically and therefore might introduce an error in a model as depicted above for θ and β-factor. Secondly, water temperature, atmospheric pressure, and electrical conductivity might provide information about other characteristics of the activated sludge process state as the following examples show. Even though a water temperature is standardized to 20 °C in the context of oxygen transfer parameters it could still provide additional information about the activity of biomass in the activated sludge tank. As another example, stormwater inflow often correlates with a sudden drop of water temperature, ambient pressure, and salinity thus changing wastewater inflow regarding soluble substances and particulate matter.

2.2.5 Activated Sludge Process Characteristics

There are numerous variations of process layout and operation of an activated sludge tank mostly depending on site-specific wastewater characteristics and treatment target. These aspects can alter the three-phase system in the activated sludge tank and thus influence the oxygen transfer. This section summarizes impacts related to operational adjustments, such as sludge retention time (SRT) and internal recirculation, and related to design aspects, such as the activated sludge tank length, upstream selectors, and primary screening.

Sludge Retention Time

The sludge retention time (SRT), also referred to as mean cell retention time (MCRT) or sludge age describes the retention time of biomass in the activated sludge reactor and is related to the treatment target. It is a function of tank volume and its biomass concentration TSS as well as all effluent flows with their respective TSS, such as waste activated sludge. Although SRT is a basic process parameter defining design, operation, and control of activated sludge systems its determination can be unreliable when WAS is withdrawn intermittently (Balbierz and Knap, 2017). In addition, a temperature based correction of SRT should be used (compare Clara et al., 2005) because biomass activity depends on activated sludge temperature. Nonetheless, SRT has been discussed in the context of aeration efficiency, as it is related to oxygen requirement and oxygen transfer efficiency. Operating an activated sludge tank at a high SRT increases oxygen requirements, whereas the advanced treatment generally increases the α-factor because of removal of readily biodegradable COD and surfactants (Gillot and Héduit, 2008; Günkel-Lange, 2013; Rosso and Stenstrom, 2006b).

SRT has been related to the α-factor because it is a key parameter that can differentiate activated sludge tank designs and treatment processes on a basic level. Rosso et al. (2005) compared results from 372 off-gas measurements on the sites of 30 WWTPs. They found a logarithmic dependency between the α-factor and a parameter that is defined by the ratio of SRT and the airflow rate for the aerobic tank volume covered by diffusers. Overall, for various diffuser types it was shown that the α-factor increases with SRT.

Reactor Type

The concentration of oxygen transfer inhibiting substances decreases with increased treatment time due to biodegradation as described in section 2.2.2. In plug flow reactors

(PFR) this results in a decrease of overall concentration of contaminants and an increase of biosorption on sludge flocs along the length of an activated sludge tank, hence, the α-factor increases in flow direction (Rosso, 2018). For example, Rosso et al. (2005) found a significant increase of α-factor in aerobic zones in flow direction of tank length in two similar activated sludge tanks operating at different SRT. The examined tank had a downstream denitrification zone. In this case no removal of oxygen transfer inhibiting substrate could occur prior to the aerated tank zones. Tapered aeration is often installed in plug flow reactors because oxygen demand is highest and α-factor is generally lowest in the influent zone. A higher diffuser density in the inlet zone than in subsequent zones provides the required oxygen to maintain stable DO in all tank zones (Baquero-Rodríguez et al., 2018).

In contrast, in continuous stirred tank reactors (CSTR) concentrations of wastewater contaminants are more uniform within the tank and also result in a uniform α-factor in different tank zones (Brade and Shahid, 1993; Rosso, 2018). Therefore, in a sequencing batch reactor (SBR) an increase of the α-factor with reaction time should be considered for the design and operation of this type of reactor (Ahmed et al., 2021b).

Treatment Processes located before aerated Activated Sludge Zones

Oxygen is transferred exclusively in aerated zones of activated sludge tanks for aerobic treatment. Treatment processes that alter the characteristics of the wastewater or the activated sludge before it is aerated in aerobic zones can positively influence the oxygen transfer efficiency. The mechanisms to improve aeration efficiency include the removal of oxygen transfer inhibiting wastewater contaminants or the biosorption on activated sludge flocs before the aerobic reactor. In particular, the removal of surfactants prior to the aeration zone has been examined in previous studies. For example, Petrovic and Barceló (2004) estimated that 90 to 95 % of overall surfactants are removed in conventional wastewater treatment as a result of adsorption to primary and secondary sludge and biodegradation in the aerobic treatment; Mohan et al. (2006) concluded that biodegradation of some surfactants is also possible under aerobic and anoxic conditions; and Garrido-Baserba et al. (2020) investigated the effect of biosorption of surfactants, colloidal, and soluble fractions on aeration efficiency in various process layouts and found an increase of surfactant removal by 27 to 56 % with biosorption configurations (contact stabilization or anaerobic selector). In this context, denitrification zones, selectors, and primary screening have been examined in other studies.

Rosso and Stenstrom (2005) demonstrated the advantages of nitrification/denitrification systems compared to conventional and nitrification systems in a comparative economic analysis. Besides the oxygen credit due to the denitrification process, a higher α-factor can be expected at higher SRT. Rosso et al. (2008) determined an increase of the α-factor from 0.37 to 0.48 to 0.59 for conventional, nitrification, and nitrification/denitrification systems. When combined, these effects overcame the additional oxygen requirements caused by a higher SRT and ultimately reduced the operating costs of aeration systems. However, it remains unclear how much of this positive effect can be attributed to the overall increase of SRT or the anoxic biodegradation of wastewater contaminants before the aerobic zone.

The latter effect has also been examined as a potential advantage of anaerobic and anoxic selectors. Selectors are implemented as upstream tanks to mix influent wastewater with return activated sludge before an activated sludge tank. Their primary function is to prevent the proliferation of filamentous bacteria. Like upstream denitrification zones, selectors could remove some readily biodegradable COD and surfactants and thus improve α-factors in subsequent aerobic activated sludge treatment. Fisher and Boyle (1999) found no improvement of the α-factor after adding a selector for test plants operated at an SRT of 7 to 10 days. Mueller et al. (2000) compared a CAS with a contact stabilization process using a selector and found an improvement of the α-factor by 10 to 15 %. Overall, Rosso et al. (2008) concluded that nutrient-removing selectors were able to further increase aeration efficiency due to the mechanisms stated above. Compared with high-rate activated sludge systems, Garrido-Baserba et al. (2020) found that contact stabilization and anaerobic selectors could improve the α-factor by 46 and 54 % based on tests in batch-scale reactors.

Similarly, enhanced carbon redirection in mechanical wastewater treatment offers the possibility to operate a subsequent activated sludge tank more energy-efficiently due to reduced oxygen requirements and increased aeration efficiency. Primary screening and filtration technologies redirect a larger share of colloidal and particulate fraction of carbon from biological treatment into the anaerobic sludge treatment process (Caliskaner et al., 2014; Franchi and Santoro, 2015; Ruiken et al., 2013). Regarding the effect on the α-factor, Pasini et al. (2020) found an improvement of aeration efficiency of 20 and 27 % when comparing screened and non-screened primary effluent in full-scale and pilot-scale, respectively. Like with upstream mechanical treatment, carbon redirection can also be enhanced with high-rate activated sludge processes (HRAS) of two-stage activated sludge systems. The expected low α-factors in the HRAS stage are subject of paper P.2.

2.2.6 Diffuser related Characteristics

For submerged aeration systems diffusers are installed on the bottom of activated sludge tanks. Designers select diffusers to meet minimum and maximum oxygen supply in the activated sludge tank. Within this specified range of demand the selection of diffusers aims to optimize energy efficiency during operation. Therefore, diffuser manufacturers optimize oxygen transfer efficiency (OTE) and diffuser pressure loss of their products. Diffuser pressure loss is defined as the pressure difference across the diffuser operated under submerged conditions. Both parameters are affected by the specifications of a diffuser and inevitable changes of diffuser properties during operation in activated sludge as described below.

Diffuser Specifications and Operation

Nowadays fine-bubble diffusers are equipped with a thin flexible membrane with fine orifices that produce bubbles with a diameter between 2 and 5 mm. In contrast coarse-bubble diffusers produce generally larger bubbles in a broader range of diameter. Higher airflow rates produce slightly larger bubbles as the membrane and its orifices are stretched more (U.S. EPA, 1989). Smaller bubbles are preferred because of the higher specific interfacial area increasing oxygen transfer efficiency. Groves et al. (1992) measured a 30 % higher OTE with fine-bubble diffusers compared with coarse-bubble diffusers. However, diffuser headloss in membranes with small orifices is generally higher and must be considered to evaluate overall energy efficiency. For most applications in conventional activated sludge systems fine-bubble diffusers are used (Baquero-Rodríguez et al., 2018).

Diffuser types for submerged aeration systems exist in various shapes such as plate, tube, and disc diffusers. Behnisch et al. (2020) compared oxygen transfer results in clean water of 65 fine-bubble diffusers and found slightly lower oxygen transfer efficiencies for disc diffusers than for tube or plate diffusers in a pilot-scale tank.

Diffuser density describes the number of installed diffusers as the ratio of total perforated area of diffusers and the bottom area of the activated sludge tank floor. Diffusers are typically distributed evenly across the tank bottom in grid layouts in each zone to avoid spiral rolls (Groves et al., 1992). In tapered aeration designs the diffuser density decreases across the tank length with decreasing oxygen requirements. However, a separate control of airflow rate by valves in each zone should still be implemented to more flexibly meet the actual oxygen demand (Åmand et al., 2013). A higher diffuser density also increases the oxygen transfer efficiency (Behnisch et al., 2020). Overall, there are diminishing returns as OTE increase is minimal at very high diffuser densities, which has to be evaluated in each individual case according to design criteria (U.S. EPA, 1989).

Diffuser Properties during Operation

During operation in activated sludge diffuser performance changes because of organic fouling and inorganic scaling phenomena. Fouling is caused by biofilm growth on the surface of diffuser membranes and in its orifices whereas scaling is caused by accumulation of inorganic precipitates such as calcium carbonate or silica inside diffuser orifices (Tchobanoglous et al., 2014; U.S. EPA, 1989). These effects generally result in an increase of pressure loss of diffusers which also increases energy demand of blowers. Aging of diffusers can also increase rigidity and hardness of membrane material thus leading to higher diffuser pressure loss (Kaliman et al., 2008; Rosso et al., 2008b; Wagner and von Hoessle, 2004). Fouling and scaling can be reduced by periodical cleaning of diffusers. Rosso (2015) suggests cleaning at least once every two years to minimize biofilm growth. Often, change of diffuser pressure loss at a certain airflow rate over time is used as a surrogate parameter to describe fouling, scaling, and aging in one parameter. To distinguish biofilm growth from aging and scaling related factors, Garrido-Baserba et al. (2016) determined DNA concentration per area of diffuser membrane by sampling and analyzing cutouts of used diffusers.

The relationship between fouling and scaling with the oxygen transfer efficiency is less conclusive. USEPA (1989) distinguish a positive relationship between OTE and diffuser pressure loss as Type I fouling from a negative relationship as Type II fouling. A positive effect of fouling and scaling on oxygen transfer efficiency can result from clogging of orifices that decreases the size of forming bubbles. On the other hand, excess biofilm growth on top of the membrane surface can hinder detachment of bubbles and result in coalescence of bubbles. Both mechanisms affect the respective bubble size and thereof resultant specific interfacial area, thus influencing OTE. Both types of fouling have been reported, e.g., Groves et al. (1992) observed a 20 % decrease of OTE of membrane diffusers after 3.5 years and, in contrast, Mueller et al. (2002) described a positive relationship between OTE and diffuser pressure loss. Both types of fouling occur but usually Type II fouling prevails which leads to a reduction of OTE (Baquero-Rodríguez et al., 2018; Garrido-Baserba et al., 2017). Some diffuser membrane materials such as silicone and PTFE+EPDM showed resistance to fouling compared to conventional materials such as EPDM and polyurethane (Rosso, 2015). The resultant effect on the fouling factor affecting oxygen transfer efficiency remained less conclusive as an increase of DNA concentration on the diffuser membrane did not always reduce the fouling factor (Garrido-Baserba et al., 2016). So far, the effect of fouling, scaling, and aging cannot be predicted in advance because they depend on diffusers, and site-specific conditions such as wastewater characteristics and maintenance efforts (Baquero-Rodríguez et al., 2018). Therefore, (Rosso et al., 2012) proposed on-site column testing of diffusers to determine performance of diffusers in situ. They suggested to conduct the testing procedure during the design and construction stage to consider site-specific effects for the selection of diffusers.

2.3 Modelling Oxygen Transfer Dynamics

Constant α-factors are, to date, a common practice in design of aeration systems worldwide. However, as outlined in the previous sections the α-factor dynamically changes in the activated sludge tank and this variance can result in failures of process design and operation (Amaral et al., 2017). Modelling these variations would be immensely useful to avoid these failures. However, at the current state of knowledge, no oxygen transfer model has been developed that can generally predict the α-factor for any kind of WWTP process layout and wastewater characteristics (Amaral et al., 2017; Baquero-Rodríguez et al., 2018). Numerous studies developed simple regression models to model the α-factor based on few input parameters. As outlined in the previous sections of chapter 2.2, oxygen transfer efficiency in the three-phase system involves many interacting mechanisms. To consider this level of complexity, models to predict oxygen transfer dynamics must also become more complex and involve more input parameters.

Table 2: Literature overview of oxygen transfer models

Source	Model prediction	Input parameters	Database/ Experimental setup	Comments
Rosso et al., 2005	α-factor and αSOTE	MCRT, airflow rate, diffuser area, diffuser submergence	Database of 372 off-gas tests on 30 full-scale WWPTs	No evidence for different α-factors for different diffuser types
Gillot and Héduit, 2008	α-factor	Equivalent contact time based on MCRT, airflow rate, diffuser submergence	Database of 27 off-gas tests on 14 full-scale WWPTs	Linear model based on equivalent contact time
Henkel et al., 2011	α-factor	SRT, MLVSS	Literature reference data and batch-scale off-gas data (88 L)	Two separate linear models based on SRT and MLVSS
Pittoors et al., 2014	k_La in activated sludge	Volume, height, diameter, (diffuser) surface area, airflow rate, diffuser submergence, bubble size, TSS	18 tests in batch-scale cylindrical tanks (3-9 L)	Dimensional analysis with k_La in clean water and activated sludge similar to Gillot et al. (2005)
Jiang et al., 2017	α-factor (dynamic prediction)	COD load	full-scale off-gas data from 3 WWTPs	Prediction of α-factor is incorporated into a model to predict the required airflow rate
Ahmed et al., 2021a	α-factor (dynamic prediction)	Soluble COD	Batch-scale SBR off-gas data (850 L)	Prediction of α-factor is incorporated into a model to predict the required airflow rate
Ahmed et al., 2021b	α-factor (dynamic prediction)	Reactor type (PFR, CSTR, step-feed PFR, MBR), soluble COD, influent COD, MLSS	Batch-scale SBR off-gas data (850 L) and simulated data from reactor models	Comparison of three separate models based on soluble COD, influent COD, and MLSS
Bencsik et al., 2022	α-factor (dynamic prediction)	SRT, influent COD, anoxic zones, diffuser depth, MLSS	Literature reference data and full-scale off-gas data	Spatial and temporal variation of α-factor is considered

Table 2 compares an excerpt of studies that included more than one input parameter to predict the α-factor (and related oxygen transfer parameters) for various WWTP processes or studies that presented approaches to dynamically predict the α-factor in real-time in activated sludge systems. As discussed in the previous sections, the current capability of oxygen transfer models is limited. Remaining knowledge gaps are outlined below:

- In practice, the oxygen transfer inhibition is not only subject to wastewater contaminants inhibiting oxygen transfer as expressed by the α-factor but also fouling and scaling processes on the diffuser membrane as expressed by the fouling factor F (see section 2.2.6). This further complicates experimental design and procedures when gradual fouling must be considered or prevented to examine a "pure" α-factor. Otherwise, analysis of oxygen transfer inhibition is often combined to an αF-factor, especially in full-scale measurements, where the individual effects cannot be separately observed. Unfortunately, many studies do not explicitly distinguish α-factor results from αF-factor results.
- Interactions between mechanisms or parameters influencing the α-factor as described in previous chapters often have not been examined before. Most studies have varied only a subset of the operating parameters or wastewater characteristics in experimental design, thus potentially overlooking effects occurring in practice. For example, standardization of oxygen transfer parameters considers the direct physical effect on the oxygen transfer process. However, a change of wastewater temperature might also correlate with other effects that additionally influence oxygen transfer (see section 2.2.5).
- The amount of variation of site-specific process layouts and wastewater characteristics that can influence oxygen transfer dynamics leads to a high complexity that models usually cannot represent. Many models are related to SRT or sum parameters that simplify influences on oxygen transfer. For example, for models related to COD influent concentration some uncertainty remains because two different wastewaters with the same COD concentration could have different oxygen transfer inhibiting effects in an activated sludge tank because the respective wastewater composition differs in concentration of surfactants or other contaminants. Consequently, models are not generally applicable and must be calibrated to adjust for site-specific conditions.
- Although the scope of laboratory analysis and online sensor measurements is extensive, its focus is to monitor the overall treatment performance of the activated sludge process instead of the aeration efficiency. As a result, the influences on oxygen transfer at the point of impact, the bubble rising in activated sludge, are largely unknown. It remains unclear whether the currently collected operating data is sufficient to properly model the oxygen transfer process or if additional

experimental data is required. For example, the use of low temporal resolution laboratory analysis data might not be suitable for real-time dynamic prediction models. It remains unclear whether the available data from online sensors and analyzers is sufficient to feed a dynamic α-factor prediction model.

Finally, it must be stated that any data describing oxygen transfer efficiency under process conditions is based on off-gas tests. Without the information about oxygen mass balance in the activated sludge reactor, no models can be developed and no conclusions about oxygen transfer dynamic can be drawn. Consequently, the implementation of off-gas measurements in full-scale applications is a requirement to advance modelling of oxygen transfer.

3 Objectives and Outline of Papers

My focus in this dissertation was to investigate the dynamics of oxygen transfer in activated sludge and the potential of oxygen transfer modelling based on long-term ex situ off-gas measurements. The dissertation is a cumulative research work with three peer-reviewed papers published in scientific journals. Although each paper answers individual research questions, the publications are linked with each other as outlined in detail below. In short, P1 and P2 comprehensively investigate the methodology of ex situ off-gas measurements and the oxygen transfer dynamics in the special process layout of two-stage WWTPs, respectively. P3 then expands on these findings and applies a machine learning approach to dynamically predict the oxygen transfer in activated sludge tanks. My combined contribution to the research field of aeration technology and prospective applications of my research findings are summarized in the final chapter 5.

Paper 1 (P1)

The paper "Determination of alpha factors for monitoring of aeration systems with the ex situ off-gas method: experience from practical application and estimation of measurement uncertainty" deals with determination of oxygen transfer parameters such as the α-factor. I used pilot scale reactors with full-scale water depth of 5.8 m that apply the ex situ off-gas method to measure oxygen transfer parameters in activated sludge for all experiments in this dissertation. This method is presented in technical guideline ASCE/EWRI 18-18 (2018). However, its description is limited to an Annex (D.1.4.4) with less than two pages of content. After more than four years of operation with continuous off-gas testing and clean water oxygen transfer comparison tests in practice, I gained operational experience with this method that is not mentioned in technical guidelines so far. The paper therefore presents limitations of the ex situ off-gas methodology and compares it with the more common in situ off-gas testing using off-gas hoods on the activated sludge tank surface. In addition, sensitivity and uncertainty analysis were performed to estimate the ex situ off-gas method's measurement uncertainty. Measurement uncertainty of the α-factor determined with the ex situ pilot plant was estimated by a theoretical error propagation approach and comparison measurements. The available information on measurement uncertainty of the off-gas method in literature was neither accurate nor comprehensive enough for the purpose of this thesis. In this regard, the experiments and results of P1 prepared the evaluation of prediction performance of a machine learning model in P3. Estimating the measurement uncertainty of the pilot plant in P1 was important to compare it with the model prediction error in P3. The paper was published in the journal Environmental Science and Pollution Research, 29, pages 87950–87968 (2022).

Paper 2 (P2)

The paper "Oxygen Transfer in Two-Stage Activated Sludge Wastewater Treatment Plants" presents results of long-term off-gas tests in two-stage activated sludge systems. Two-stage WWTPs are an interesting research object because operation differs tremendously from CAS systems which also affects the aeration efficiency. Conditions in a high-rate activated sludge tank with short retention times and high sludge loading produce a wastewater-sludge-matrix that drastically inhibits oxygen transfer. Nonetheless, the increased biosorption of wastewater contaminants in this stage can result in an energy-efficient operation of biological wastewater treatment under certain conditions. Optimizing operation and control strategies for the two-stage process recently sparked new interest in the research community. But no comprehensive study of oxygen transfer dynamics in two-stage activated-sludge was available, yet. I collected off-gas data over a period of 13 months on the site of a two-stage WWTP treating municipal wastewater (1.35 Mio. PE). Based on these long-term experiments, loading cases of the α-factor for the static design of aeration systems in two-stage systems were defined. These complement α-factor loading cases in German technical guideline DWA-M 229-1 (2017) to design aeration systems. In addition, the influences on oxygen transfer inhibition in the two activated sludge stages were discussed based on their different operation. For example, the fate of surfactants in the two-stage process and their effect on oxygen transfer inhibition was elucidated. For WWTP operators, the change of α-factor during rainy and dry weather was discussed as well as the potential of reverse flexing as a maintenance technique to reduce diffuser fouling in the respective treatment stages. Collecting and analyzing extensive off-gas data for a two-stage system in P2 was especially relevant for P3, because the presented methodology to develop oxygen transfer models included an unusual WWTP process layout. Otherwise, it would have remained unclear whether the prediction models developed in P3 were suitable for process layouts other than the conventional systems. The paper was published in the journal Water 2021, 13(14), 1964.

Paper 3 (P3)

The paper " Dynamic alpha factor prediction with operating data - a machine learning approach to model oxygen transfer dynamics in activated sludge" presents a novel approach to model oxygen transfer in activated sludge. In the last decades, many studies examined specific influences on oxygen transfer as presented in chapter 2.3. However, no conclusive and generally applicable models were defined to model oxygen transfer dynamics so far. Many influencing parameters are superimposed and specific to a WWTP's wastewater characteristics. Most models to dynamically predict α-factors are based on typically discussed influences on spatial and temporal variation of the α-factor in the activated sludge tank, such as COD influent concentration, total suspended solids, and sludge retention time. These models still require laborious parameterization to consider site-specific parameters of full-scale processes and laboratory analysis. Furthermore, focusing on a few influencing parameters might miss the full potential of available operating data. Nowadays, monitoring of activated sludge tanks with in-situ sensors and ex situ analyzers collects operating data with high temporal resolution. Nonetheless, it was unclear if operating data typically available to WWTP operators is sufficient to describe the complex mechanisms involved with the oxygen transfer in a three-phase system as outlined in chapter 2.2. However, there is an abundance of online sensor data at large WWTPs that has not been utilized to its full potential to develop oxygen transfer models before. After long-term off-gas measurements collecting extensive datasets on the sites of three different WWTPs, I was able to apply a data-driven approach to develop oxygen transfer models. For the first time, a supervised machine learning approach was used to dynamically predict α-factors with predictor variables based exclusively on operating data available to WWTP operators. The results for four different activated sludge stages were presented and the limitations of the black box machine learning approach was discussed. The paper was published in the journal Water Research (Volume 231, 1 March 2023, 119650).

4 Publications

Overview of Papers

Paper 1 Schwarz, M.; Trippel, J.; Engelhart, M.; Wagner, M. (2022)

Determination of alpha factors for monitoring of aeration systems with the ex situ off-gas method: experience from practical application and estimation of measurement uncertainty

Environ Sci Pollut Res, 29, pages 87950–87968 (2022)

https://doi.org/10.1007/s11356-022-21915-2

Paper 2 Schwarz, M.; Behnisch, J.; Trippel, J.; Engelhart, M.; Wagner, M. (2021)

Oxygen Transfer in Two-Stage Activated Sludge Wastewater Treatment Plants

Water 2021, 13(14), 1964

https://doi.org/10.3390/w13141964

Paper 3 Schwarz, M.; Trippel, J.; Engelhart, M.; Wagner, M. (2022)

Dynamic alpha factor prediction with operating data - a machine learning approach to model oxygen transfer dynamics in activated sludge

Water Research, Volume 231, 1 March 2023, 119650

https://doi.org/10.1016/j.watres.2023.119650

P.1 Determination of alpha factors for monitoring of aeration systems with the ex situ off-gas method: experience from practical application and estimation of measurement uncertainty

Authors: Schwarz, M.; Trippel, J.; Engelhart, M.; Wagner, M.

Keywords: Activated sludge, Aeration, α-Factor, Measurement uncertainty, Oxygen transfer, Sensitivity analysis

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P.1.1 Abstract

Performance of aeration systems in wastewater treatment plants (WWTP) under process conditions can be monitored with off-gas tests. The ex situ off-gas method transfers activated sludge from an adjacent aeration tank into aerated columns to determine oxygen transfer parameters (e.g., the α-factor). This method is an alternative to in situ off-gas testing with hoods at the tank surface; however, its application and measurement uncertainty have not been examined yet. We outline our experience from long-term off-gas testing with two pilot-scale test reactors (8.3 m^3 volume). Global variance-based sensitivity analysis using Sobol' indices revealed oxygen concentration in off-gas and dissolved oxygen as the most important input quantities to determine α-factors accurately. Measurement uncertainty of other instruments was negligible. These findings are transferable to in situ off-gas hoods because the methods are similar. Random measurement error of α-factors was estimated with uncertainty analysis and comparison measurements to a relative standard deviation of about ± 2.8 % for our ex situ pilot setup. Diffuser fouling, biofilm growth, or sensor drift caused systematic errors avoidable by maintenance. Additional mixing of bubble column due to sludge inflow into ex situ tanks led to a systematic overestimation of α-factors at lower airflow rates. Hence, the ex situ off-gas method is not suitable to determine α-factors for the design of aeration systems but offers unique possibilities for research of oxygen transfer dynamics and development of aeration equipment because ex situ columns can be operated independently from a full-scale activated sludge tank.

P.1.2 Introduction

Aeration is an energy-intensive process in activated sludge (AS) biological wastewater treatment. Measurement of oxygen transfer parameters in activated sludge tanks is essential for design and operation of aeration systems. Clean water testing is an established method to determine oxygen transfer performance of diffusers (ASCE 2–06 2007; EN 12255–15 2003). Still, wastewater treatment plant (WWTP) operators face a decline of oxygen transfer under process conditions in activated sludge tanks. This is caused by inhibitory effects of wastewater and activated sludge components in the soluble and solid phase as well as the impact of fouling, scaling, and aging of diffusers resulting in poor bubble formation and rise as reviewed by Baquero-Rodríguez et al. (2018). The α-factor summarizes these oxygen transfer inhibiting effects as the ratio of oxygen transfer in process water to clean water. Design and operation of aeration systems must consider oxygen transfer in process conditions, which can be measured with off-gas methods (ASCE 18–18, 2018; DWA-M 209 2007).

P.1.2.1 Off-gas testing in wastewater treatment

Off-gas testing methods have been used for numerous applications in design and operation of aeration systems as well as research of gas transfer in activated sludge, as the following examples show. In several studies, off-gas tests were used to examine impacts on oxygen transfer by activated sludge characteristics or WWTP operation and process layout (Leu et al. 2009; Rosso et al. 2008, 2005; Schuchardt et al. 2005). Studies of this type allow to model aspects of the oxygen transfer. For example, Jiang et al. (2017) proposed a dynamic model to predict α-factors based on the relationship between the α-factor and chemical oxygen demand (COD). Off-gas tests can be part of the design process of the aeration system. Rosso et al. (2012) performed on-site testing of various diffusers to determine the influence of process specific wastewater properties on oxygen transfer (and pressure loss) during design phase to enable more accurate design of aeration systems. Off-gas tests can also be utilized to monitor the operation of a WWTP. Trillo et al. (2004) applied off-gas hoods for a feed-forward dissolved oxygen (DO) control to reduce aeration energy costs. Leu et al. (2010) measured oxygen and carbon dioxide transfer rates to predict effluent ammonia. Hellinga et al. (1996) already argued that in contrast to selective point measurements with sensors, off-gas measurements in treatment plants with covered aeration tanks could be a worthwhile addition to liquid phase analysis to monitor the overall biological treatment process. This application of off-gas testing could also be combined with monitoring of emissions in the future. Myers et al. (2021) measured dissolved and off-gas nitrous oxide (N_2O) in a conventional activated sludge (CAS) WWTP and estimated volumetric mass transfer coefficient of nitrous oxide based on the mass transfer coefficient for oxygen. Baeten et al. (2021) used off-gas analysis to detect several emissions (CO_2, CH_4, N_2O) in an aerobic granular sludge WWTP. So far, off-gas analyses have been used in numerous studies, but application of gas analyzers is not part of the typical instrumentation on WWTPs yet.

P.1.2.2 Comparison of off-gas methods

The off-gas method with off-gas hoods on the surface of aeration tanks is first described by Redmon et al. (1983) and explained in ASCE/EWRI 18–18 (2018) and DWA-M 209 (2007). It allows to measure oxygen transfer efficiency from which an oxygen uptake rate (OUR) can be calculated based on a dissolved oxygen mass balance. Boyle et al. (1989) demonstrate the possibilities of off-gas measurements for OUR online monitoring without the necessity of error-prone ex situ batch OUR respirometry devices. The Redmon Engineering Company used ex situ off-gas column tests to determine α-factors in the 1980s. The method is first described by Rieth and Polta (1987) and included in ASCE 18–18, section D.1.4.4. We refer to the method as ex situ column off-gas testing. It is an alternative to in situ off-gas hoods that allows to examine oxygen transfer in activated sludge transferred from an adjacent AS tank into a separate column. The off-gas measurement is therefore independent from the operation of the activated sludge tank and its aeration system. Both the in situ off-gas hood and the ex situ column method allow to determine the same oxygen transfer parameters, e.g., standard oxygen transfer rate (SOTR), standard oxygen transfer efficiency (SOTE), OUR, and the α-factor. However, the application differs in certain aspects of the methodology and operation.

Placing multiple off-gas hoods to cover an activated sludge tank is generally less expensive than using ex situ columns to reach the same coverage. Determination of an overall α-factor of the process design is especially relevant in plug-flow reactors, tapered aeration or tanks with varying oxygen concentrations in different tank areas (Rosso et al. 2005; Stenstrom et al. 2006). Additionally, more sensors and flow measurements are necessary with the ex situ off-gas measurement compared to off-gas hoods which increases maintenance effort. In situ off-gas hoods are more convenient for a WWTP operator to monitor an installed aeration system's performance over long periods or estimate it with single off-gas test series. However, a variation of the oxygen transfer cannot be attributed distinctly to either activated sludge related (α) or fouling related (F) causes. In contrast, ex situ columns allow to mitigate fouling by regular cleaning of diffusers and therefore distinguish the α-factor from the fouling factor. In addition, clean water testing is mandatory to determine the α-factor and easier to perform in ex situ columns than in a full-scale AS tank. Overall, an ex situ column allows to change certain properties of the aeration system and operation without interfering with the WWTP operation. Applications for research purposes could include varying tank geometry (especially blow-in depth), changing diffusers to find suitable types for a certain application, examining the effect of maintenance methods (e.g., reverse flexing, high-pressure cleaning, and acid injection) on reinstating pressure loss of diffusers, or performing spiking experiments to change wastewater characteristics and study the resulting effect on oxygen transfer. While off-gas hoods can only be placed in aerated zones, the ex situ method allows to transfer sludge from non-aerated zones into the column and determine oxygen transfer parameters. This also allows to determine oxygen transfer parameters for activated sludge in tanks without submerged aeration systems or covered tanks where hoses for sludge transfer can be installed unlike off-gas hoods. This represents a unique advantage of the ex situ off-gas method to research potential emissions from non-aerated zones in the future.

P.1.2.3 Estimation of measurement uncertainty

Technical guidelines such as ASCE 18–18 or DWA-M 209 define measurement models to determine oxygen transfer parameters, e.g., oxygen transfer rate (OTR), oxygen transfer efficiency (OTE), or the α-factor. These measurement models define a functional relationship between several input quantities recorded by sensors or instruments and an output quantity, e.g., the α-factor as a measurand. Currently, guidelines are missing a stochastic component that considers measurement uncertainty of instruments recording input quantities. Instead, guidelines propose a measurement uncertainty to be expected for results if the applied method was conducted according to standard. For the use of in situ off-gas hoods DWA-M 209 (2007) estimated a measurement uncertainty of ± 5 to 10 % for SOTR in activated sludge depending on tank shape and size. ASCE 18–18 (2018) referred to a comparison of several methods by Capela et al. (2004) and Mahendraker et al. (2005) and concluded that the examined methods estimated oxygen transfer parameters in activated sludge within 10 to 15 % of each other depending on the examined method. Redmon et al. (1983) originally reported a reproducibility of ± 10 % for OTE with in situ off-gas tests in activated sludge which primarily depended on changing conditions at a sampling point rather than accuracy of the analytical system.

None of the studies examined the ex situ column off-gas method. In addition, it remains unclear which measured input quantity is most important when determining oxygen transfer parameters or the α-factor. Another detail that is often accepted without further revision is the use of correction terms for standardization. For off-gas methods β and θ correction factors are applied to consider the effect of salts on the effective oxygen saturation and temperature on the oxygen transfer, respectively. These empirically determined correction terms are also estimates of quantities which are known imperfectly and could vary between applications of off-gas tests (Stenstrom and Gilbert 1981). Sensitivity analysis is a method to examine these issues. Its principle is to identify the effect of changes or uncertainty of input quantities on the model output (Turányi 1990). Variance-based methods such as Sobol's method (Sobol' 1993) aim to explain the effect of variance in model inputs on variance in model outputs. The thereby calculated sensitivity indices distinguish first-order and total effects indices. A first-order index represents the influence of an individual input quantity on the variance of the model output quantity. A second-order index explains interactions between two input quantities on the model output which cannot be explained by the sum of their first-order effects. Total effects indices summarize all higher-order indices (including second-order and above) to represent the total impact of an input quantity on output variance (Homma and Saltelli 1996). Sensitivity indices are represented by values between 0 and 1. When comparing sensitivity indices of input parameters, a higher value indicates a stronger influence of the input quantity on the model output. It is therefore more important to define or measure accurately to yield reasonable results. The methodology of sensitivity analysis (SA) using Sobol' indices is described, e.g., in Saltelli et al. (2004) or Sobol' and Kucherenko (2005).

P.1.2.4 Objectives

Our study describes the setup and operation of the ex situ column off-gas method in-depth and thereby complements information missing in technical standards. In addition, there are three objectives to improve future applications of the ex situ column off-gas method: (1) we determine the most influential input quantities for determination of α-factors according to ASCE 18–18 with a sensitivity analysis, (2) we estimate the method's random and systematic measurement error, and (3) we discuss causes of these errors and other constraints of the ex situ column off-gas method.

P.1.3 Methods

P.1.3.1 Design and operation of ex situ columns

Pilot-scale test reactors were used to determine oxygen transfer parameters applying an off-gas method described in Appendix D.1.4.4 of ASCE/EWRI 18–18 (2018). The method is a variant of the steady-state oxygen uptake rate (OUR) technique, where OUR is measured within the ex situ columns with off-gas analysis instead of an additional respirometry device. Figure P.1.1 shows a flow diagram of the process for one ex situ aeration tank. Our pilot plant featured two aeration tanks with duplicate machinery and instruments to examine two AS tank zones in parallel.

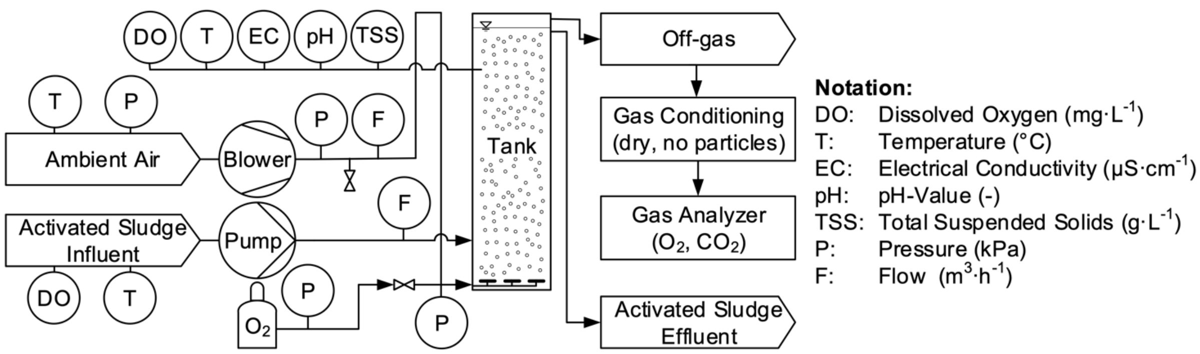

Figure P.1.1 Flow diagram of an ex situ setup for steady-state off-gas measurements

Tank dimensions were 1.2 m × 1.2 m × 5.8 m (L × W × H) with a volume of 8.3 m^3. The tank height was chosen to resemble typical water depths of AS tanks and therefore bubble rise conditions in the column. Columns were equipped with fine-bubble disc diffusers with a diffuser density of 13.5 % (ELASTOX-T EPDM TYP B, WILO GVA, Wülfrath, Germany). Oil-free rotary vane vacuum blowers (CB.40, D.V.P. Vacuum Technology spa, Italy) were controlled by frequency converters to set airflow rates (specified for aerated tank volume − $q_{Vol,aer}$) between 0.5 and 2.5 $Nm^3 \cdot m^{-3} \cdot h^{-1}$. Airflow rate was standardized (101.325 kPa, air temperature of 0 °C, dry air) and measured with thermal mass flowmeters (t-mass 150, Endress + Hauser AG, Reinach, Switzerland) in the inflow only. As with in situ off-gas hoods, this assumes that inert gas constituents such as nitrogen are conservative within the reactor and therefore net transfer of these gases is negligible. Pressure in air pipes (Cerabar PMC21, Endress + Hauser AG, Reinach, Switzerland) was measured after blowers and before diffuser distribution frame to determine pipe pressure loss and diffuser pressure loss. Sludge transfer pumps (AGNM02 NEMO®, NETZSCH Holding, Selb, Germany) pumped AS from a nearby AS tank through DN 100 hoses into the columns at the height of the disc diffusers. Depending on the examined AS tank zone a hose length of up to 100 m was installed and the maximum transfer time of the AS to the test columns was 90 s. Sludge flow was measured with electromagnetic flowmeters (Promag W 400, Endress + Hauser AG, Reinach, Switzerland) and transfer pumps controlled by frequency converters to set a constant hydraulic retention time (HRT) of 15 min as recommended by ASCE/EWRI 18–18. Effluent sludge was directed in free flow through a DN 150 hose from an overflow edge back into the nearby AS tank downstream of sludge intake.

Determining oxygen transfer parameters of AS in the columns required further sensors and instruments for measurement. Atmospheric pressure (Cerabar PMC21, Endress + Hauser AG, Reinach, Switzerland), atmospheric temperature (Omnigrad T TST434, Endress + Hauser AG, Reinach, Switzerland), and electrical conductivity in AS (Indumax CLS50D, Endress + Hauser AG, Reinach, Switzerland) were measured for standardization of oxygen transfer parameters (20 °C water temperature, 101.325 kPa atmospheric pressure, 1.000 $mg \cdot L^{-1}$ total dissolved solids). Off-gas concentrations of oxygen (paramagnetic sensor) and carbon dioxide (NDIR) were recorded with a gas analyzer (X-STREAM Enhanced, Emerson Electric Co., MO, USA) that received dry off-gas free of particles from a gas conditioning unit (CSS-V, M&C TechGroup, Ratingen, Germany). Off-gas was collected from the sealed column hood. To quickly monitor changing process conditions, a low hood height of 0.2 m above water surface was implemented on top of the columns. Depending on airflow rate setting the mean gas sample residence time in the hood was between 2 and 4 min, which included off-gas transport from hood to gas analyzer. Foaming could complicate off-gas collection in low hoods. Thus, the pilot plant was equipped with a U-shaped off-gas pipe that withheld foam from off-gas collection.

Sensors were cleaned twice a week to prohibit biofilm growth and solids deposition affecting optical instruments and calibrated as necessary. Because of its relevance for the off-gas method, two-point calibration of the off-gas analyzer was performed twice a week using calibration gases with 5 % CO_2, 16 % O_2, and 100 % N_2 for zero point. Potential biofilm build-up on the reactor tank walls was prevented with monthly cleaning and visual inspection to ensure only suspended biomass transferred from the adjacent full-scale AS tanks was examined in the ex situ reactors for off-gas measurements.

P.1.3.2 Data recording and processing

A suitable interval for data compression must be short enough to record changes in WWTP operation or wastewater composition that could affect oxygen transfer and long enough to produce distinguishable datapoints for further analysis. Depending on the response time of equipped sensors in a pilot plant, determining α-factors in intervals of a few minutes is possible. A typical measurement period for off-gas testing is 30 min to 2 h (ASCE 2018). From the recorded data, a mean α-factor and a dispersion coefficient (e.g., standard deviation) is determined to estimate uncertainty of measurement or steady-state conditions. In continuous ex situ measurements these α-factors form a time series that describes the change of oxygen transfer in the continuous stirred tank reactor (CSTR). However, the determined oxygen transfer parameter or α-factor does represent not only the oxygen transfer of the sludge inflow at that moment but also of the previously transferred activated sludge already in the column. Therefore, determination of α-factors in an ex situ CSTR requires longer intervals depending on hydraulic retention time (HRT) and the resulting residence time distribution of the activated sludge in the columns. From our experience sufficient mixing was provided by aeration in the columns. An airflow rate of 2.2 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (0.38 $Nm^3 \cdot m^{-3} \cdot h^{-1}$), which is a commonly used design criterion to maintain solids in suspension (Water Environment Federation, 2018), was exceeded during off-gas testing. Additionally, a constant lateral flow of activated sludge transferred into the tank potentially mixed dead space beneath the diffuser distribution frame. Unless a sensor drift occurred, DO sensors showed the same DO concentration in the reactors. Therefore, ideal mixing conditions within the columns can be assumed and the residence time distribution (t) in a single ideal CSTR can be expressed as a cumulative distribution function as

$$F_{(t)} = 1 - e^{-\frac{t}{HRT}} \qquad (P.1.1)$$

Based on this ideal relationship, in our pilot plant, 63 % of activated sludge transferred into the test column was exchanged within the HRT of 15 min. Accordingly, after 30, 45, and 60 min, 86 %, 95 %, and 98 % of sludge were replaced. As a result, a 1-h interval is a suitable interval for data compression for an ex situ reactor operated at an HRT of 15 min to determine α-factors.

To maintain steady-state conditions within a selected interval of data compression, some parameters (i.e., reactor influent flow, influent DO, DO in reactor, oxygen uptake rate, and oxygen transfer rate) should remain constant to determine oxygen transfer parameters (Boyle 1983). Therefore, ex situ columns allow to control reactor inflow and internal DO. Influent DO is steady if the examined activated sludge tank is controlled to a DO setpoint. However, oxygen uptake rate and oxygen transfer rate depend on wastewater composition and operation of AS process. Both are variable throughout a longer measurement period. Consequently, a test period to determine α-factors must be long enough to collect data sufficiently and short enough to keep steady-state conditions.

In our setup, data was recorded in 30-s intervals by online sensors and compressed as 1-h averages. This results in high resolution data that can detect variations within the diurnal cycle of WWTP operation. It also prevents autocorrelation of measured values and converts the collected time series data to resemble a cross-sectional dataset. From our experience, the required constant conditions as described above were met within a 1-h interval unless airflow rate or DO setpoint were changed manually or according to a schedule within an interval.

P.1.3.3 Determination of oxygen transfer parameters

Determination of the α-factor and other oxygen transfer parameters is based on the well-established equation for actual oxygen transfer rate under process conditions (AOTR) which represents the transfer of oxygen without any standardization in activated sludge (United States Environmental Protection Agency 1989). The equation contains several factors to consider the influence of wastewater characteristics and varying ambient conditions during off-gas testing. Rearranged for the α-factor it is expressed as:

$$\alpha = \frac{AOTR}{F \cdot k_L a_{cw,20} \cdot (\beta \cdot \tau \cdot \Omega \cdot C_{20}^{*} - C_{(t)}) \cdot \theta^{T-20} \cdot V} \qquad \text{(P.1.2)}$$

In off-gas measurements, AOTR ($g \cdot h^{-1}$) is calculated from the oxygen transfer efficiency (OTE) at a certain airflow rate. ASCE 18–18 describes how OTE is calculated from a mass balance of inlet and outlet oxygen and carbon dioxide concentrations measured with an off-gas analyzer. It also defines dimensionless standardization parameters to calculate standard oxygen transfer rate (SOTR) where τ is the oxygen saturation ratio at operating temperature and at 20 °C, Ω is the oxygen saturation pressure correction factor for 101.325 kPa, β is the ratio of oxygen saturation in process water and clean water, and θ is the temperature correction coefficient for water temperatures of 20 °C. Although ASCE 18–18 provides a general description of the ex situ column method, we provide further explanations based on practical experience below and added all equations to determine the α-factor in Appendix 1 of this paper.

Fouling factor — F (−)

The fouling factor is defined as the ratio of oxygen transfer performance of used and new diffusers. During long-term off-gas measurements in activated sludge, diffuser performance is reduced because of scaling, fouling, and aging of diffusers. Ex situ columns could be used to specifically determine the fouling factor F if diffusers were not cleaned periodically. Significant increases of fouling measured by pressure loss are rare within the first three months without maintenance (Rosso 2015; Rosso et al. 2012). On the other hand, ex situ columns allow to maintain diffuser performance and therefore to determine α-factors with minimal impact of fouling if maintained properly within shorter intervals. To mitigate fouling, regular pressure cleaning and reverse flexing of diffusers and acid addition into air pipes can be performed (Odize et al. 2017; Rosso 2018; Wagner and Stenstrom 2014). Because our objective was to determine oxygen transfer as α-factor instead of αF-factor, reverse flexing was performed twice a week and membrane surface of diffusers was cleaned with high pressure once a month. A previous study has shown that the effect of fouling during long-term off-gas measurements could be kept low when applying this maintenance (Schwarz et al. 2021). Here, clean water tests repeated over a period of 13 months revealed a decrease of SOTR of 2 to 6 % depending on airflow rate and a dynamic wet pressure increase of about 1 kPa. For even longer periods, an exchange of diffusers seems advisable.

Clean water testing — $k_La_{cw,20}$

Clean water (cw) testing is required to determine the denominator of the α-factor which is based on the linear relationship between airflow rate and SOTR in clean water. We used different probes in clean water and process water because clean water tests required faster dissolved oxygen (DO) probes than off-gas measurements at high airflow rates. Electrochemical DO probes (Oxymax COS51D, Endress + Hauser AG, Reinach, Switzerland) with a fast response time t_{90} of 30 s were used. Slower optical DO probes Oxymax COS61D, Endress + Hauser AG, Reinach, Switzerland) produced similar results but at lower accuracy. These were used in process conditions as long-term testing did not require a fast response time and their lower maintenance allowed more reliable operation in activated sludge. Furthermore, off-gas measurements were performed at a steady sludge inflow, while non-steady-state clean water tests were not. Consequently, differences of bubble rise and gas holdup in the columns could have occurred between test methods as discussed later. A steady-state clean water test is neither described in technical guidelines nor practically feasible at the setup’s scale.

Oxygen saturation concentration — C^*_{20}

Steady-state off-gas cannot provide an estimate of effective oxygen saturation concentration C^*_{20} which would result in the activated sludge at zero respiration rate. Therefore, it was estimated by a mid-depth model also considering influence of temperature and pressure (i.e., τ, Ω) (compare with Jiang and Stenstrom 2012). However, the effect of soluble total dissolved solids (TDS) as estimated by the β-factor cannot be determined in continuous off-gas testing. Instead, it is estimated from electrical conductivity by a conversion factor of 2 $mg \cdot L^{-1}$ TDS/3 $\mu S \cdot cm^{-1}$ (see DWA-M 209, 2007).

Volume V

Volume of tanks should be measured accurately because it directly affects SOTR. Clean water and off-gas testing should be conducted with the same water volume to prevent a systematic error.

α-Factor

To determine the α-factor in the aeration tank, the airflow rate in the columns has to be adjusted to set DO in the ex situ columns within the range of DO in the examined aeration tank (ASCE 2018; Boyle 1983). This operation preserves steady-state conditions of DO and aims to reproduce the gas transfer found in the aeration tank as close as possible in the ex situ test column. In this case the setup resembles the in situ off-gas hood method, provided that the same diffuser type, diffuser density, and tank depth are implemented as in the examined aeration tank.

P.1.3.4 Sensitivity analysis of ex situ off-gas measurements

ASCE 18–18 gives little information about measurement uncertainty of the off-gas method. It remains unclear which input quantity is most important to produce accurate results. The principle of sensitivity analysis (SA) is to identify the effect of changes of input quantities on the model output (i.e., the α-factor) (Turányi 1990). Examined input quantities to determine the α-factor as described above were off-gas oxygen ($O_{2,e}$) and carbon dioxide concentrations ($CO_{2,e}$), water temperature (T_w), dissolved oxygen (DO and $C_{(t)}$), electrical conductivity (EC) of the AS, atmospheric pressure (p_{atm}), and the airflow rate (q_{air}). In the underlying model to determine α-factors, some input quantities are correlated (especially $O_{2,e}$, $CO_{2,e}$, DO), e.g., higher $CO_{2,e}$ values generally correlate with lower $O_{2,e}$ values. The model is non-additive because input quantities interact with each other. This means that changing two inputs has a different effect on the output than the sum of their individual effects which must be considered in sensitivity analysis (Saltelli et al. 2004). Instead of simulating this dependency in the input quantities during the sampling process with individual models, we collected results of long-term measurements in a conventional activated sludge (CAS) WWTP with 700,000 population equivalent treating municipal wastewater over a period of 11 months. The resulting dataset contains 10,700 recorded α-factors as 1-h intervals. In this dataset distribution of input, quantities represent typical operation of a CAS plant including seasonal variations and therefore cover the range of input quantities required for a global sensitivity analysis (Saltelli et al. 2004; Sudret 2007).

Applying the methodology of sensitivity and uncertainty analysis, we examined the following aspects of α-factor determination with the ex situ off-gas method:

Method 1: Examine the individual influence of measured input quantities

An elementary "one factor at a time" (OAT) analysis only considers the relationship between the output and the variation of one individual input quantity around one baseline case where all other input quantities are kept at their nominal values (Saltelli 1999). This local method would be restricted to one observation of input quantities at a time (baseline case) to determine the α-factor. To consider the range of input quantities, we performed the analysis for our whole dataset and reported average deviations of the α-factor. The results were generated by varying all observations of a specific input quantity by ± 1.0 % and ± 5.0 % from their nominal values (baseline cases) and recalculating the average α-factor of the dataset. The baseline cases are the input quantities and corresponding α-factors as determined by ASCE 18–18 (2018) from our dataset. Relative percentage change of this value and the average α-factor of the dataset was calculated for comparison of variations of all input parameters. In elementary OAT, any interactions of input quantities are discounted. Nonetheless, this elementary OAT analysis can be performed if the variation of input quantities is small (Saltelli 1999; Saltelli et al. 2019). The small variation of ± 1.0 % and ± 5.0 % is chosen to represent typical measurement uncertainties of the input quantities.

Method 2: Examine the individual influence of correction factors

We applied the same method as in Method 1 and varied the correction factors θ and β as well as the conversion factor for TDS/EC according to their ranges found in literature.

Method 3: Estimate measurement uncertainty of our setup

We performed an uncertainty analysis to estimate the measurement uncertainty to expect when determining α-factors with our ex situ off-gas columns. The measurement uncertainty of the α-factor was affected by the measurement uncertainty of all instruments involved to measure input quantities. A common approach is to use a derivative based method for error propagation to determine a combined standard uncertainty (Joint Committee for Guides in Metrology 2008). However, this uncertainty would only be valid locally for an individual measurement and does not consider the distribution of errors. To take these aspects into account, we estimated the uncertainty for all measurements in our dataset by the following steps: To create a base for comparison, all observations of recorded input quantities and thereof determined α-factors in our dataset were regarded as "true" values, i.e., reference quantity values. Random measurement error of instruments was simulated by sampling 4000 values of every input quantity according to the instrument's individual measurement uncertainty for every observation in our dataset (n = 10,700). A detailed overview of a priori instrument measurement uncertainties and their distributions which are specific to our pilot setup is listed in Appendix 2 of this paper and technical information of each instrument is also provided by manufacturers online. Most measurement uncertainties were chosen according to technical information by the manufacturer. However, because optical sensors for measurement of dissolved oxygen were operated in activated sludge the uncertainty of ± 1 % of reading stated by manufacturer was considerably lower than our own measurements. Therefore, we assumed an uncertainty of ± 0.1 $mg \cdot L^{-1}$ (uniform distribution) ± 5 % of reading (± SD, normal distribution) as described in Appendix 2. In total, 42 million theoretical α-factors were determined based on the instrument measurement uncertainty that represented the expected uncertainty of the α-factors defined as "true" values. Finally, theoretical α-factors were compared with the measured "true" α-factors in our dataset.

Method 4: Examine the individual influence of measured input quantities in our setup

Sobol' sensitivity indices were determined in a global sensitivity analysis. The global SA estimated the output uncertainty due to the uncertainty of individual input quantities or combinations thereof. Sobol' indices were calculated from a decomposition of the output's variance. The aim was to identify the impact of input quantities on measurement uncertainty of α-factors for our specific pilot plant. As in Method 3, the results are based on the specific measurement uncertainties related to the instruments and sensors used in our pilot plant (see Appendix 2) and illustrate the importance of all input quantities' measurement uncertainty when performing off-gas tests with the ex situ method. The general concept is described in Saltelli et al. (2004) and first introduced by Sobol' (1993). We used a Monte Carlo estimation of Sobol' indices with improved formulas of Jansen (1999) and Saltelli et al. (2010) to determine first-order and total effects Sobol' sensitivity indices. A practical application of this SA can be found in Jadun et al. (2017), who compared variance-decomposition methods on a real model and evaluated it as most suitable to determine total effects indices.

Statistics and visualization were done using R 3.6.3 (R Core Team 2020), tidyverse package (v1.3.0) for visualization (Wickham et al. 2019), data.table package (v1.14.0) for data handling (Dowle and Srinivasan 2021) and sensitivity package (v1.26.1) to perform sensitivity analysis (Iooss et al. 2021).

P.1.4 Results and discussion

First, we discuss the results of the sensitivity analysis to point out theoretical causes of measurement uncertainty. Afterwards, we present our results from a direct comparison of α-factors measured simultaneously in two pilot reactors from the same AS zone. Based on this, we discuss possible causes of random and systematic error affecting the ex situ off-gas method's measurement uncertainty.

P.1.4.1 OAT sensitivity analysis of α-factor determination

An average α-factor of 0.70 was calculated according to ASCE 18–18 for our whole dataset of measured input quantities. Table P.1.1 displays the relative change from this average when all observations of an individual input quantity were adjusted by ± 1 % or ± 5 %, see Method 1. The mean value ± standard deviation (SD) of all input quantities is listed to characterize the dataset underlying the analysis.

Table P.1.1 OAT sensitivity analysis: relative percentage change of mean α-factor for adjusted input parameters

Input parameter	Mean ± SD	α-factor: rel. perc. change (%) calculated with input quantities adjusted by			
		- 5 %	- 1 %	+ 1 %	+ 5 %
O_2 in off-gas ($O_{2,e}$)	17.9 ± 0.7 %	+ 31.2	+ 6.3	- 6.3	- 31.9
Water temperature (T_w)	18.4 ± 2.9 °C	+ 1.8	+ 0.4	- 0.4	- 1.7
Atmospheric pressure (p_{atm})	1,013 ± 9 hPa	+ 1.3	+ 0.3	- 0.2	- 1.2
Dissolved oxygen (DO)	2.1 ± 1.4 $mg \cdot L^{-1}$	- 1.2	- 0.3	+ 0.3	+ 1.3
CO_2 in off-gas ($CO_{2,e}$)	2.2 ± 0.4 %	+ 0.7	+ 0.1	- 0.1	- 0.7
Vol. spec. airflow rate (q_{air})	1.5 ± 0.3 $Nm^3 \cdot m^{-3} \cdot h^{-1}$	+ 0.5	+ 0.1	- 0.1	- 0.5
Electrical conductivity (EC)	1,380 ± 360 $\mu S \cdot cm^{-1}$	+ 0.1	0.0	0.0	- 0.1

The input quantities are sorted by descending absolute influence on the α-factor determination. When nominal values of O_2 in off-gas were reduced by 5 % across all measured observations, the mean α-factor increased by 31.2 % based on the mean α-factor of 0.70. In contrast, a decrease of electrical conductivity by 5 % increased α-factor negligibly by 0.1 %. The exact relative percentage changes obtained by the analysis depend on the underlying dataset. The OAT sensitivity analysis confirms that the oxygen concentration in the off-gas is by far the most influential input quantity to determine the α-factor. Thus, maintenance and calibration of the gas analyzer is essential for off-gas testing. Adjusting water temperature, atmospheric pressure, and dissolved oxygen by up to ± 5 % had similar impacts on the average α-factor. This theoretical approach ignores the fact that each sensor recording these input quantities has a different measurement uncertainty. Errors of more than ± 5 % are common for airflow meters or DO sensors when used in AS. Additionally, the closer water temperatures were to 20 °C the lower the relative percentage change of α-factor, because of its standardization to 20 °C.

In Method 2, the same approach is applied to analyze the impact of correction factors for standardization. Inexact values of theses constants could be an additional source of measurement uncertainty (Joint Committee for Guides in Metrology 2008). Table P.1.2 lists the relative or absolute percentage changes from the average α-factor of the dataset for variations of three standardization correction factors as input quantities.

Table P.1.2 OAT SA: relative/absolute percentage change of mean α-factor for adjusted standardization factor

Standardization factor		**Adjustments of nominal value (-) Rel./abs. change of α-factors (%)**				
θ temp. correction factor (theta)	Input (-)	1.008	1.02	1.024	1.028	1.047
	Abs. perc. change of α-factors (%)*	4.2	1.1	-	1.1	6.3
TDS/EC conversion factor	Input (-)	0.55	0.62	0.67	0.72	0.90
	Rel. perc. change of α-factors (%)	+ 0.2	+ 0.1	-	- 0.1	- 0.3
β-factor (beta)	Input (-)	0.9	0.95	0.991	0.994	0.998
	Rel. perc. change of α-factors (%)	- 9.2	- 4.1	-	+ 0.3	+ 0.7

* Absolute percentage change of α-factor was determined for variations of θ because deviations changed from positive to negative (and vice versa) at water temperatures of 20 °C.

The temperature correction factor θ applies a geometric correction to standardize mass transfer of oxygen to 20 °C. It is set to 1.024, but the empirically determined factor attempts to combine several effects such as changes in diffusivity of oxygen, viscosity, or surface tension. Reported values range from 1.008 to 1.047 (Stenstrom and Gilbert 1981), while ranges from 1.020 to 1.028 are reasonable according to US Environmental Protection Agency (1989). As theta is influenced by turbulence, it depends on the type of aeration system. Changing theta to a different factor requires support of substantial data (Stenstrom and Gilbert 1981). Within the range of 1.024 ± 0.004, the average α-factor of our dataset deviated by 1.1 %. However, temperature correction becomes more influential for off-gas measurements at more extreme temperatures than present in our dataset (18.4 ± 2.9 °C).

The conversion from total dissolved solids to electrical conductivity is $2\ mg\cdot L^{-1}$ TDS = $3\ \mu S\cdot cm^{-1}$ (DWA 2007), see Appendix 1, equation A5. The factor 0.67 is confirmed by Behnisch et al. (2021) who determined an average of 0.7 for various salts. AWWA standard methods list a broader range between 0.55 and 0.9 (AWWA, 2017).

However, the effect on α-factor determination is negligible (below ± 0.3 %) at the salt concentrations expected in municipal wastewater. In this case, an adjustment of the β-factor also has low impact on the resulting α-factors. In our dataset, a mean β-factor of 0.991 was determined by equation A5. Uncertainty about the correct estimate of the β-factor in municipal wastewater remains as Eckenfelder et al. (1956) report β as approximately 0.95 and ASCE 18–18 states that it can vary from 0.8 to 1.0, but is generally close to 1.0. An adjustment of β to 0.95 or 0.9 results in a relative change of α-factor of − 4.1 % and − 9.2 %, respectively. If equation A5 did not consider the effect of salts on effective oxygen saturation concentration correctly, it would directly impact the α-factor. For certain industrial (and possibly municipal) wastewaters, this could introduce a systematic error when determining the α-factor.

P.1.4.2 Variance-based sensitivity analysis of the ex situ off-gas method

The OAT sensitivity analysis described before did not consider possible interactions of input quantities on the α-factor and only selectively compared importance of input quantities for fixed variations of ± 1 % and ± 5 %. Sobol' indices based on variance decomposition detected interactions of input quantities and considered their differing measurement uncertainties (see Method 4). The resultant first-order and total effects Sobol' sensitivity indices for all input quantities are sorted in descending importance from left to right in Figure P.1.2. Boxplots visualize the distribution of indices for all 10.500 samples of the underlying dataset instead of adding bootstrap confidence intervals for every index.

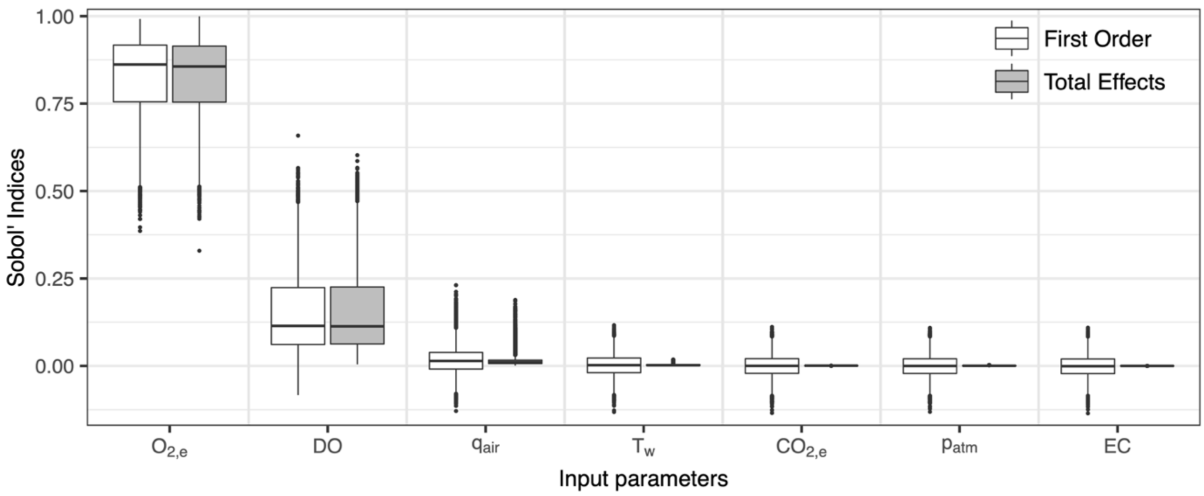

Figure P.1.2 Comparison of first order and total effects Sobol' sensitivity indices for input quantities to determine α-factors: oxygen in off-gas ($O_{2,e}$), dissolved oxygen (DO), airflow rate (q_{air}), water temperature (T_W), carbon dioxide in off-gas ($CO_{2,e}$), atmospheric pressure (p_{atm}), and electrical conductivity (EC)

Oxygen concentration in off-gas ($O_{2,e}$) has the highest first-order Sobol' index followed by dissolved oxygen (DO). These Sobol' indices show the influence of each input quantities' measurement uncertainty on the variance of the output (i.e., the α-factor).

Compared with the previous OAT sensitivity analysis (see Table P.1.1), this confirms the importance of oxygen concentration in the off-gas whereas the impact of dissolved oxygen is higher than before. Hence, we can conclude that accurate measurement of oxygen in off-gas and dissolved oxygen must be prioritized for reliable off-gas testing with the instruments used in our pilot setup. In contrast, all other input quantities have low first-order and total effects indices which means that their measurement uncertainty had a negligible effect on the uncertainty of the α-factor.

The output was primarily influenced by first-order effects because these were equal to total effects. Although the model is non-additive, no significant interactions were present when sampling input quantities within their measurement uncertainty. Otherwise, total effects indices would be larger than first-order indices. Interactions were present in the model although larger deviations of input quantities were tested (data not shown). On average, sum of first-order and total effects Sobol' indices were close to 1, which also confirms that interactions between input quantities were negligible. Some first-order Sobol' indices were negative for the less influential input quantities. This was not caused by correlated input quantities because sampling was random. Negative first-order Sobol' indices can occur when the sample size is insufficient (Glen and Isaacs 2012) or when output is not distributed normally (Menberg et al. 2016). Nonetheless, indices can be assumed zero because they were distributed evenly around zero. Although the first-order indices could be not as robust as under perfect conditions, they still demonstrate a distinct difference in the input quantities' importance as discussed above.

P.1.4.3 Random measurement error of ex situ off-gas tests

We simulated random measurement error of our setup based on the input quantities' individual measurement uncertainty as described in Method 3. The resulting difference of the "true" α-factors and sampled α-factors produce random measurement errors to estimate the measurement uncertainty across a dataset of long-term measurements. The average α-factor and its standard deviation was 0.70 ± 0.025 (relative standard deviation of ± 3.7 %).

Next, we estimated the measurement uncertainty of our setup by considering measurement error from comparison measurements. Our dataset included periods where both pilot reactors were operated at the same airflow rate and hydraulic retention time while transferring AS from the same aeration zone. In total, 1400 pairs of simultaneously determined α-factors collected at 1-h intervals provided a direct comparison to estimate the pilot setup's measurement uncertainty. This direct comparison of two identically equipped and operated ex situ off-gas columns was a suitable method to estimate the method's measurement uncertainty because no activated sludge with a known α-factor can be used for calibration. The results are shown in Figure P.1.3.

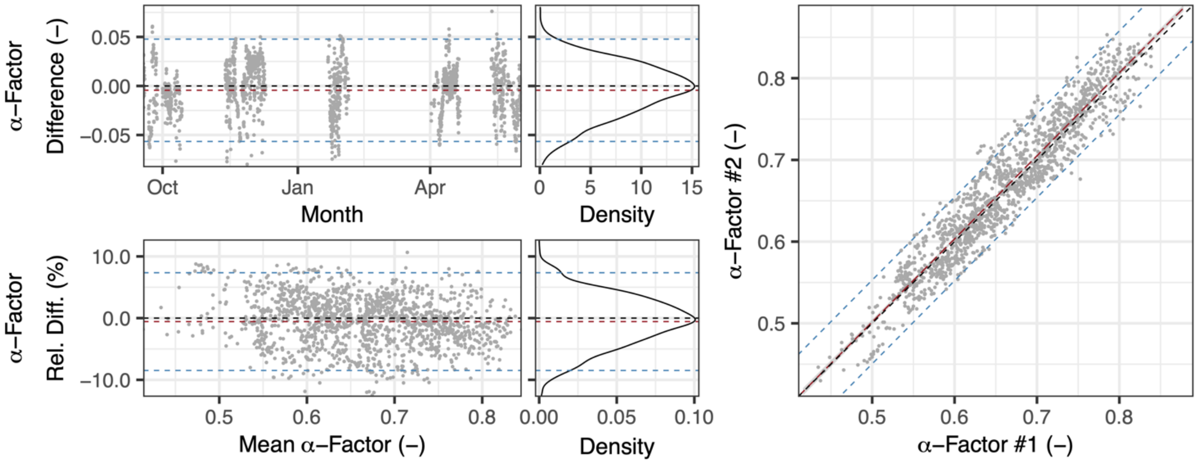

Figure P.1.3 Comparison of α-factors simultaneously determined in two ex situ off-gas columns

Figure P.1.3 is divided into five separate diagrams. The upper diagrams display the difference of α-factor #1 and α-factor #2 over a long-term measurement period (left) and their resulting distribution (center). The lower counterparts show the relative difference (ratio of difference and common mean of both α-factors) for the mean α-factor of both setups (left) and the resulting distribution (center). The right diagram directly compares each pair of α-factors. Dashed lines represent the ideal case without any difference (black), the average of all observations of our dataset (red) and ± 2 SD around the mean or the corresponding 95 %-prediction interval (blue). This comparison of two pilot setups shows that individual measurements of α-factors approximately follow a normal distribution with a SD of ± 4 % of relative difference, whereas long-term testing provides more consistent results. The average of all observations (red dashed line) is close to the ideal case (black dashed line) with a relative difference lower than 1 %.

The measurement error equals random measurement error plus systematic measurement error. As discussed in the next section, systematic measurement errors could not be excluded or corrected as the offset shifted between measurement periods. Figure P.1.3 thus visualizes spread and distribution of random and systematic error values. Based on these the measurement uncertainty of the individual setups was estimated. One assumption therefore is the normal distribution of the observed differences of α-factors (compare Figure P.1.3, center top) and the differences of α-factor #1 and #2 with the ideal case. This assumption is common for random error of measurements. If the distribution is the same for both setups their mean (μ) and standard deviation (σ) can be derived from:

$$\mathcal{N}(\mu_{obs}, \sigma_{obs}^2) = \mathcal{N}(\mu_1, \sigma_1^2) - \mathcal{N}(\mu_2, \sigma_2^2) \tag{P.1.3}$$

with:

$\mathcal{N}(\mu_{obs}, \sigma_{obs}^2)$:
Normal distribution of the observed (measured) differences of α-factors as shown in Figure P.1.3 with mean $\mu_{obs} = \mu_1 - \mu_2 = -0.0044 \approx 0$ and standard deviation $\sigma_{obs} = 0.026$

$\mathcal{N}(\mu_{1,2}, \sigma_{1,2}^2)$:
Normal distribution of the individual setups determining α-factor #1 and #2.

Because standard deviations for the individual setups (σ_1 and σ_2) were assumed identical they can be calculated as follows:

$$\sigma_{obs}^2 = \sigma_1^2 + \sigma_2^2 = 2\sigma_{1,2}^2 \tag{P.1.4}$$

$$\sigma_{1,2} = \sqrt{\frac{\sigma_{obs}^2}{2}} = 0.018 \tag{P.1.5}$$

While the difference of means μ_1 and μ_2 was close to zero, the standard deviations σ_1 and σ_2 were calculated from Eq. (P.1.5) as 0.018 for α-factors in the dataset of the comparison. At an average α-factor of 0.66 in the dataset, the mean relative standard deviation was ± 2.8 %. The lower left diagram in Figure P.1.3 shows that relative difference of α-factors does not change significantly at lower or higher α-factors, which indicates that the relative standard deviation can estimate measurement uncertainty across the whole range of possible α-factors. Table P.1.3 compares these measurement error results of measured values from two pilot setups with the results simulated with the uncertainty analysis (see Method 3).

Table P.1.3 Comparison of measurement uncertainty based on simulation and measurements of ex situ off-gas tests

Data source and analysis	**Simulated in uncertainty analysis**	**Parallel measurement in two ex situ pilot reactors**
Number of observations in dataset	n = 10.500	n = 1.400 (in each reactor)
Mean α-factor of dataset	0.70	0.66
Mean standard deviation (-)	± 0.025	± 0.018
Mean relative standard deviation (%)	± 3.7	± 2.8

The average values of standard deviation and relative standard deviation are similar for both approaches. However, data analyzed from parallel measurement in two ex situ pilot reactors was compressed to 1-h intervals. In contrast, the values simulated in the uncertainty analysis represented the random error of measurement expected for instrument readings within the overall response time of all instruments. This period is shorter than one hour and cannot be determined exactly for our setup because response time of instruments varied, and gas sampling was dependent on airflow rate. The values simulated in the uncertainty analysis overestimate uncertainty, because more than one distinct measurement could take place within 1 h and thereby decrease the overall random measurement error.

Repeating off-gas measurements is generally recommendable due to random measurement errors. Our results show that a considerable measurement error could remain if only a single 1-h interval of an α-factor was determined. Repeated long-term measurements can compensate for this as the following example illustrates: In the case of our pilot reactors, a single measurement of an α-factor of 0.66 would be associated with a relative standard deviation of ± 2.8 %. Repeating this measurement 5, 10, or 20 times would decrease relative standard deviation to 1.3 %, 0.9 %, or 0.6 %, respectively. This example is valid under the assumption of a pure random measurement error. However, a systematic measurement error influencing multiple observations in sequence could still have a larger impact than demonstrated here.

P.1.4.4 Systematic measurement error of ex situ off-gas tests

Comparative measurements were performed in five distinct periods of more than 10 days that are separated in Figure P.1.4 The titles of the individual diagrams state the period of measurement, average relative difference (%), and the number of recorded 1-h intervals (n) within that period (maintenance excluded). Diagrams on top present the distribution of relative difference with a rug marking individual datapoints while diagrams on the bottom show the direct comparison of each pair of α-factors. As in Figure P.1.2, dashed lines indicate the ideal case of no deviation (black) the mean within that period (red) and ± 2 SD around the mean or the 95 %-prediction interval (blue).

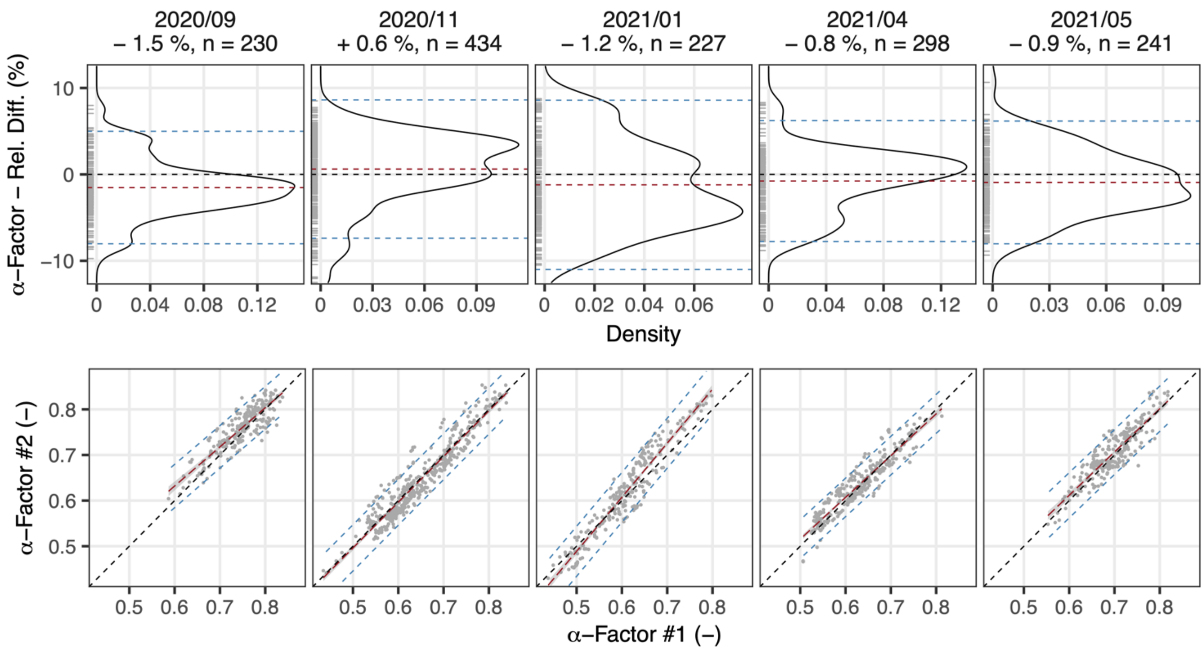

Figure P.1.4 α-factor comparison split into five off-gas measurement periods

The comparison shows that the distribution of relative differences and their average varied between measurement periods. Relative differences were not always normally distributed. This suggests that the ex situ off-gas measurement was subject to systematic measurement errors that changed between or within measurement periods. The systematic measurement error as a relative difference was within a range lower than ± 1.5 % in our setup. It is worth mentioning that this cannot identify systematic measurement errors occurring at the same time and evenly in both setups. Therefore, a systematic measurement error could be higher than the reported relative difference of ± 1.5 %.

Systematic measurement error could be caused, among other reasons, by fouling of diffusers, biofilm growth, sensor drift, or imperfect clean water testing and therefore reduced by proper maintenance of the setup and extensive clean water testing. Based on our data, systematic measurement error could not be quantitatively attributed to potential causes as discussed below. Consequently, an unknown systematic measurement error cannot be corrected when estimating the measurement uncertainty. The comparison in Figure P.1.4 and thereof derived relative difference of up to ± 1.5 % could not conclusively distinguish random measurement error from systematic measurement error. Nonetheless, it exemplarily demonstrates the effect and acknowledging the potential causes listed below may help to minimize systematic measurement errors when performing ex situ off-gas measurements.

Fouling, scaling, and aging of diffusers affects the oxygen transfer performance of an aeration system. Odize et al. (2017) found that reverse flexing helped to reduce pressure loss during operation but did not improve fouling factor effectively. Therefore, the membrane surface of diffusers was cleaned with high pressure before the individual measurement periods to mitigate fouling. Within the long-term off-gas testing period of 11 months, pressure loss increased on average by 2 kPa for both pilot reactors, but pressure loss and relative deviations of α-factors were not correlated.

Preventing excessive biofilm growth within the reactors is critical. Sessile biomass in the ex situ columns increases overall oxygen respiration and therefore alters DO concentrations and oxygen driving force in the columns when compared to suspended biomass in the AS tank. Consequently, ex situ columns could fail to accurately measure oxygen transfer conditions in an AS tank because of this systematic error. Reactor tank walls were cleaned regularly concurrently with diffusers to prevent this effect. Overall, no significant biofilm production was observed during testing. However, the impact of biofilm growth remains an unquantifiable source of error.

Sensor drift of off-gas analyzer or DO sensors could result in a systematic measurement error. A small drift of oxygen concentrations in the off-gas would have a disproportionate effect on the α-factor as shown by the sensitivity analysis. Regular calibration depending on the gas analyzer's requirement is advisable. Outliers in collected data could be identified a posteriori by large drifts marked in a calibration protocol. Moreover, biofilm growth on DO sensors submerged in AS affected their accuracy and required regular cleaning. A duplicate or triplicate measurement is advisable as it allows to identify outliers of single defective sensors a posteriori. Once these outliers were detected and removed from our dataset, no correlation with the relative difference of the α-factor was apparent. From our experience, other sensors and instruments involved in the measurement were less error-prone. Details on implementations in our setup are stated in the "Methods" section.

Clean water testing results are the denominator of the α-factor. Results of linear regression equations (SOTR ~ q_{air}) were similar for both reactors, but deviations were more probable at extreme airflow rates. Extensive clean water testing beyond the usual range of set airflow rates is advisable. Nonetheless, at airflow rates below 0.5 $Nm^3 \cdot m^{-3} \cdot h^{-1}$ accuracy of the airflow meter was insufficient in our setup. Once α-factors determined at low airflow rates were excluded, no correlation with the relative deviation between pilot reactors was apparent.

Water volume directly affects determination of oxygen transfer parameters and should be kept constant during testing as described in the "Methods" section. In our setup, no deviation related to differences of tank volume was expected because of the identical geometry of aeration columns.

P.1.4.5 Limitations of the ex situ off-gas method

Unlike in situ measurements with off-gas hoods at the surface of an aeration tank, oxygen transfer in the AS is examined ex situ with the method discussed here. Sludge transfer from an AS tank and aeration in a column with a different geometry than the AS tank could skew the α-factors determined with ex situ off-gas measurements under certain conditions.

Firstly, the positioning of sludge transfer hoses in the AS tank limits the zone that can be examined with the ex situ columns. In situ off-gas hoods are similarly restricted to cover small areas of an aeration tank. In the case of insufficient mixing in the AS tank, sludge characteristics at the sampling point could result in an undetected error. Therefore, sludge transfer hoses should be positioned in a sufficiently mixed zone.

Secondly, during transfer of aerated AS in sludge transfer hoses additional oxygen is dissolved from the gas phase while oxygen consumption of the biomass reduces it. In our dataset, oxygen transfer rates in the ex situ columns were on average $71 \pm 16\ g \cdot m^{-3} \cdot h^{-1}$ and oxygen uptake rates were similar at $68 \pm 17\ g \cdot m^{-3} \cdot h^{-1}$. However, it remains unclear whether the two opposing effects were balanced in the sludge transfer hoses. It is possible that more oxygen is dissolved than consumed under turbulent flow conditions in the hoses, which would result in an overestimation of the α-factor. Therefore, hose lengths should either be as short as possible or of the same length to reduce a potential systematic measurement error. In our application, hose lengths of up to 100 m were used.

Thirdly, the sludge transfer into the column produces a lateral flow at the height of the diffusers that is only present during off-gas testing, not during clean water testing. Figure P.1.5 shows the relationship of α-factor and volume specific airflow rate for our setup where off-gas testing was performed during dry weather in the same aeration zone of a CAS WWTP. Off-gas tests were performed at a constant sludge inflow with a HRT of 15 min so that turbulence in the columns was only influenced by airflow rate. The same data is depicted as individual data points with a local polynomial regression fit as a dashed line (left) and boxplots (right).

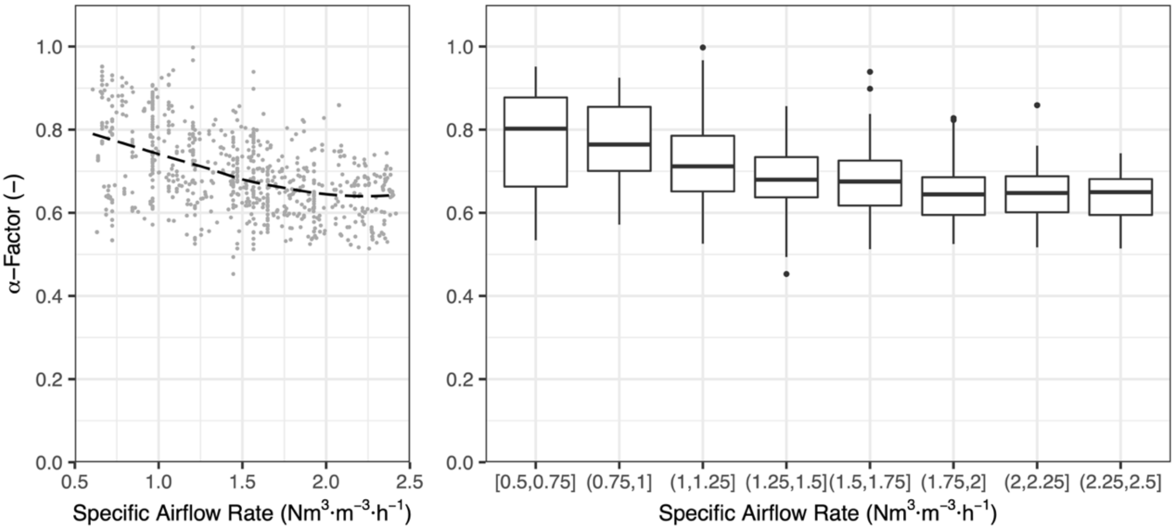

Figure P.1.5 α-factors at specific airflow rates in the ex situ column at constant sludge inflow

Both diagrams show that α-factor increases at lower airflow rates. During off-gas testing in AS, oxygen transfer is improved by higher turbulence as the rising bubble plume is additionally mixed by the sludge inflow. Consequently, a systematic overestimation of α-factors is possible, especially at low airflow rates where gas–liquid ratio is particularly low. The effect can be reduced by setting higher airflow rates that create a similarly high turbulence in off-gas and clean water testing. Nonetheless, this systematic error is setup specific and should be quantified for each ex situ column. Although Figure P.1.5 suggests that a further decrease of α-factor is limited at high airflow rates, the “true” α-factor in the AS tank is difficult to determine with the ex situ method. Nonetheless, an off-gas measurement with in situ off-gas hoods is preferable if α-factors are determined to design the aeration system of the examined AS tank.

Fourthly, determination of standard aeration efficiency (SAE) relies on accurate measurement of power consumption of blowers. Blowers equipped in a pilot-scale ex situ setup cannot accurately represent the power consumption of aeration in a full-scale AS tank. In contrast, in situ off-gas measurements with off-gas hoods use the blowers of the AS tank and should therefore be preferred to determine SAE.

P.1.5 Conclusions

Below, we summarize our findings about the application of ex situ column off-gas testing and its measurement uncertainty to determine α-factors in activated sludge tanks.

- We determined the most important input quantities of the ex situ off-gas method with a "one factor at a time" (OAT) sensitivity analysis and a global variance-based sensitivity analysis using Sobol' indices. The analysis was based on measurement uncertainties of required instruments and revealed that oxygen concentration in off-gas was the most important input quantity to determine oxygen transfer parameters (e.g., the α-factor). It was followed by dissolved oxygen concentration because its measurement in activated sludge could be unreliable. The uncertainties of all other input quantities were negligible.
- We performed an uncertainty analysis for a dataset of long-term measurements based on the measurement uncertainties of instruments in our pilot setup and estimated measurement uncertainty of the α-factor as a relative standard deviation of about ± 3.7 %. A direct comparison of α-factors from parallel operation of ex situ pilot reactors under the same conditions transferring AS from the same aeration zone resulted in a similar relative standard deviation of about ± 2.8 %. This value represents the measurement uncertainty of a single value recorded with the ex situ off-gas method. The theoretically determined relative standard deviation of ± 3.7 % and the relative standard deviation of ± 2.8 % determined from practice in our pilot setup are lower than a measurement uncertainty of ± 5 to 10 % estimated in literature before. Thus, a more accurate off-gas measurement seems possible. We recommend estimating the measurement uncertainty of α-factors theoretically for the installed instruments when planning an ex situ pilot setup as shown in Method 3. In any case, repeating measurements is advisable to produce more accurate results and reporting a measurement uncertainty of the method is beneficial to interpret results. Nonetheless, systematic measurement errors can be present and caused, e.g., by fouling of diffusers, biofilm growth, sensor drift, or imperfect clean water testing. In our experience, systematic measurement errors of about ± 1.5 % of α-factor can be caused by these issues which can rarely be identified a posteriori and only reduced by proper maintenance of the setup.
- The α-factor is standardized with correction factors to consider the influence of temperature and total dissolved solids on oxygen transfer according to standard guidelines. OAT sensitivity analysis revealed that impact of correction factors on the α-factor was lower than measurement uncertainty of the most important input quantities (oxygen concentration in off-gas and activated sludge). However, temperature correction factor θ became increasingly important when off-gas testing was conducted in activated sludge at water temperatures deviating from 20 °C. Because θ was empirically estimated as 1.024, an unknown systematic measurement

error could result when comparing oxygen transfer results from tests at significantly different temperatures. The influence of salts on the effective saturation concentration as represented by the β-factor was estimated with a formula that has negligible effect on α-factor. Nonetheless, for off-gas tests in AS treating industrial wastewater with high salt contents the β-factor should be validated by additional tests to avoid a systematic measurement error.

- In general, the findings for the ex situ off-gas method are transferable to in situ off-gas hoods because the same instruments are used to determine the α-factor. We outlined systematic influences that differentiate the methods from each other, such as changes of oxygen balance in inflow or higher turbulence in ex situ columns due to sludge transfer. We conclude that the ex situ method is not suitable to determine α-factors to design aeration systems because a systematic overestimation of α-factor at low airflow rates is probable. In contrast, off-gas hoods are suitable to monitor oxygen transfer in activated sludge tanks, e.g., for compliance testing, because resulting α-factors represent in situ conditions. In addition, full coverage of tanks is less expensive and operation easier to maintain than with ex situ reactors. However, the possibility to operate ex situ reactors independently from AS tanks offers unique possibilities for research of oxygen transfer dynamics in AS and development of aeration equipment. It could see a future application in the parallel measurement of oxygen transfer and greenhouse gas emissions (such as nitrous oxide) in aerated and non-aerated zones.

P.1.6 Appendix

P.1.6.1 Equations to determine the α-factor from measured input quantities

This annex contains all equations used to determine the α-factor with the ex situ method. Most notable are the adjustments made to allow a determination of the α-factor based on online sensor data (estimating TDS from electrical conductivity and calculating $C_{S,T,St}$ from a polynomial). With these a reader can replicate the sensitivity analysis discussed above with own data.

MR_i: Molar ratio of oxygen to inert substances

$$MR_i = \frac{\frac{O_{2,in}}{1-CO_{2,in}}}{1-\frac{O_{2,in}}{1-CO_{2,in}}} = 0.265 \qquad \text{(A1)}$$

with:

$O_{2,in}$: Inlet oxygen concentration of 20.946 %

$CO_{2,in}$: Inlet carbon dioxide concentration of 0.0407 %

$C_{S,T,St}$: Oxygen saturation concentration at water temperature Tw ($mg \cdot L^{-1}$)

$$C_{S,T,St} = \frac{2234.34}{(T_w+45.93)^{1.31403}} \qquad \text{(A2)}$$

with:

T_w: Water temperature (°C)

ASCE 18-18 refers to tabulated values by Benson and Krause Jr, (1984), the polynomial above is defined in DWA-M 229-1 and calculates these values (DWA, 2017).

$C_{S,md}$: Oxygen saturation concentration at mid-depth and standard conditions ($mg \cdot L^{-1}$)

$$C_{S,md} = 9.09 \cdot \frac{h_D}{2 \cdot 10.35} \qquad \text{(A3)}$$

with:

h_D: Blow-in depth (m)

Mid-depth saturation model based on DWA-M 209 (DWA, 2007), the mid-depth model is also recommended in Jiang and Stenstrom (2012). The effective saturation depth is setup-specific and was about 50 % of blow-in depth in our setup, which was determined in clean water tests by comparison with oxygen saturation at the surface according to DWA-M 209 (2007).

OTE_f: Oxygen transfer efficiency under process conditions (%)

$$OTE_f = \frac{MR_i - \left(\frac{O_{2,e}}{1-O_{2,e}-CO_{2,e}}\right)}{MR_i} \tag{A4}$$

with:

$O_{2,e}$: Oxygen concentration in off-gas (%)

$CO_{2,e}$: Carbon dioxide concentration in off-gas (%)

The parameter is calculated depending on the gas analyzer output and/or off-gas conditioning. For other variants the reader is referred to ASCE 18-18 (2018) or DWA-M 209 (2007).

β: β-factor (beta) (-)

the ratio of oxygen saturation in process water to clean water at equivalent conditions of water temperature partial pressure

$$\beta = 1.00 - 0.01 \cdot \frac{TDS}{1000} \quad \text{or} \quad \beta = 1.00 - 0.01 \cdot \frac{\frac{2}{3} \cdot EC}{1000} \tag{A5}$$

with:

TDS: Total dissolved solids ($mg \cdot L^{-1}$)

EC: Electrical conductivity ($\mu S \cdot cm^{-1}$)

Online monitoring requires continuous measurement of TDS. It is therefore approximated with the electrical conductivity. A common conversion is 2,000 $mg \cdot L^{-1}$ TDS = 3,000 $\mu S \cdot cm^{-1}$ (DWA, 2007).

C^*_{20}: Standardized effective oxygen saturation at process conditions ($mg \cdot L^{-1}$)

$$C^*_{20} = C_{S,md} \cdot \tau \cdot \Omega = C_{S,md} \cdot \frac{C_{S,T,St}}{9.09} \cdot \frac{p_{atm}}{101.325} \tag{A6}$$

with:

τ: Temperature correction (tau) of effective saturation concentration (-)

Ω: Pressure correction (omega) of effective saturation concentration (-)

p_{atm}: Atmospheric pressure (kPa)

$OTE_{sp,20}$: Oxygen transfer efficiency per unit of driving force at std. conditions (%/mg·L^{-1})

$$OTE_{sp,20} = \frac{OTE_f}{C_{20}^* - C_{(t)}} \cdot \theta^{20-T_w} \quad \text{(A7)}$$

with:

$C_{(t)}$: Dissolved oxygen concentration in the ex situ column (mg·L^{-1})

θ: Temperature correction factor (theta) = 1.024 (-)

$SOTE_{pw}$: Standard oxygen transfer efficiency under process conditions (%)

$$SOTE_{pw} = OTE_{sp20} \cdot C_{20}^* \cdot \beta \quad \text{(A8)}$$

$SOTR_{pw}$: Standard oxygen transfer rate in process water (g·h^{-1})

$$SOTR_{pw} = q_{air} \cdot 299.3 \cdot SOTE_{pw} \quad \text{(A9)}$$

with:

q_{air}: Airflow rate, e.g., volume specific (Nm^3·m^{-3}·h^{-1})

α: α-factor (alpha) (-)

Ratio of k_La_{pw} in process water to k_La_{cw} in clean water at equivalent conditions of tank geometry, mixing, etc.

$$\alpha = \frac{SOTR_{pw}}{SOTR_{cw}} \quad \text{(A10)}$$

with:

$SOTR_{cw}$: Standard oxygen transfer rate in clean water (g·h^{-1}) measured at the same airflow rate q_{air} as in process water, see also ASCE 2-06 (2007), EN 12255-15 (2004) or DWA-M 209 (2007). It is linearly dependent on the airflow rate and can therefore be calculated from a setup-specific linear regression model.

P.1.6.2 Measurement uncertainty of input quantities to determine α-factors

Below the measurement uncertainty of all sensors and instruments is listed that were used in the uncertainty analysis and sensitivity analysis. If not specified otherwise a normal distribution is assumed with a coverage factor of 1 (± 1 SD).

$O_{2,e}$ – Oxygen concentration in off-gas

Paramagnetic sensor X-STREAM Enhanced, XEGP-C-14-B40-0-B40-0-O26-0-O26-0-000-0-3-0-8-0-0-1-0-0-B-B (Emerson Electric Co., Missouri, USA)

± 0.5 % of reading (repeatability, confirmed by own measurements at ambient air)

$CO_{2,e}$ – Carbon dioxide concentration in off-gas

Nondispersive infrared sensor X-STREAM Enhanced, XEGP-C-14-B40-0-B40-0-O26-0-O26-0-000-0-3-0-8-0-0-1-0-0-B-B (Emerson Electric Co., Missouri, USA)

± 0.5 % of upper range limit of 5 % (repeatability, confirmed by own measurements in ambient air)

DO - Dissolved oxygen $C_{(t)}$

Digital, optical measurement of dissolved oxygen based on fluorescence quenching COS61D-1009/0-AAA1A4 (Endress+Hauser AG, Reinach, Switzerland)

According to technical information by the manufacturer measurement uncertainty is stated as 0.01 $mg \cdot L^{-1}$ or ± 1 % of reading. However, this accuracy recorded under laboratory calibration conditions is far from the readings we measured in activated sludge. Even under laboratory conditions Helm et al. (2018) found a drift of ± 0.2 $mg \cdot L^{-1}$ of optical sensors within 1 month of use. Näykki et al. (2013) conservatively estimated uncertainty of measurement at ± 0.15 $mg \cdot L^{-1}$ (± SD) for reference DO measurements in interlaboratory comparison for several types of DO sensors. Based on own measurements with two or three sensors operated in the same reactor column (though at different depth of submergence) we defined a reasonable measurement uncertainty of optical DO sensors in activated sludge as ± 0.1 $mg \cdot L^{-1}$ (uniform distribution) ± 5 % of reading (± SD, normal distribution). This is a conservative estimate because the mean value of two or three sensors is used to determine the α-factor.

T_w – Water temperature

Temperature probe in digital dissolved oxygen sensor based on fluorescence quenching COS61D-1009/0-AAA1A4 (Endress+Hauser AG, Reinach, Switzerland)

± 0.75 % of reading (uniform distribution, based on own measurements with two or three sensors operated in the same reactor column, no information provided by manufacturer)

EC - Electrical conductivity in activated sludge

Toroidal potentiometric conductivity sensor Indumax CLS50D -10M7/0-AA1B31 (Endress+Hauser AG, Reinach, Switzerland)

± (5 $\mu S \cdot cm^{-1}$ + 0.5 % of reading) (maximum measured error, a uniform distribution is applied for sampling)

p_{atm} - Atmospheric pressure at blower air intake

Absolute and gauge pressure Cerabar PMC21-21W0/0-AAl U2KBWBJA (Endress+Hauser AG, Reinach, Switzerland)

± 0.3 % of upper range limit of 2 bar (maximum measured error, a uniform distribution is applied for sampling)

q_{air} – Airflow rate

Thermal mass flow meter t-mass A 150-14D9/0-6AAB15 (Endress+Hauser AG, Reinach, Switzerland)

± 4 % of reading (uncertainty increases at airflow rates below 6 $Nm^3 \cdot h^{-1}$ that were not set)

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P.2 Oxygen Transfer in Two-stage Activated Sludge Wastewater Treatment Plants

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Keywords: aeration; alpha (α); fine-bubble diffusers; high-rate activated sludge systems (HRAS); off-gas; reverse flexing; surfactants; TOC F/M ratio; TOC sludge loading; wastewater treatment

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P.2.1 Abstract

Aeration is an energy-intensive process of aerobic biological treatment in wastewater treatment plants (WWTP). Two-stage processes enable energy-efficient operation, but oxygen transfer has not been studied in depth before. In this study, α-factors were determined with long-term ex situ steady-state off-gas measurements in pilot-scale test reactors (5.8 m height, 8.3 m^3) coupled to full-scale activated sludge basins. A two-stage WWTP with more than 1 Mio population equivalent was studied over 13 months including rain and dry weather conditions. Operating data, surfactant concentrations throughout the two-stage process, and the effect of reverse flexing on pressure loss of diffusers were examined. The values of α_{mean}, α_{min}, and α_{max} for design load cases of aeration systems were determined as 0.45, 0.33, and 0.54 in the first high-rate carbon removal stage and as 0.80, 0.69, and 0.91 in the second nitrification stage, respectively. The first stage is characterized by a distinct diurnal variation and decrease in α-factor during stormwater treatment. Surfactants and the majority of the total organic carbon (TOC) load are effectively removed in the first stage; hence, α-factors in the second stage are higher and have a more consistent diurnal pattern. Proposed α-factors enable more accurate aeration system design of two-stage WWTPs. Fouling-induced diffuser pressure loss can be restored effectively with reverse flexing in both treatment stages.

P.2.2 Introduction

Aeration is an essential process in aerobic biological wastewater treatment. In most wastewater treatment plants (WWTPs), it accounts for more than half of the net energy consumption (Baquero-Rodríguez et al., 2018; Reardon, 1995; Rosso et al., 2011). Engineers rely on technical standards providing design guidelines to properly design aeration systems (DWA, 2017; United States Environmental Protection Agency, 1989;

Water Environment Federation, 2018). Various WWTP process configurations are possible depending on wastewater composition and required effluent target. Each process configuration demands individual design considerations for the aeration system. Technical guidelines, therefore, provide α-factors to consider inhibiting effects on oxygen transfer in the activated sludge (AS). The α-factor determines oxygen transfer efficiency as the ratio of oxygen transfer under process conditions compared to clean water. However, comprehensive research on oxygen transfer in two-stage AS processes is not available. This study provides planners with α-factors required for the design of aeration systems in a two-stage configuration. We discuss the impact of stormwater treatment and fluctuations of operating parameters such as TOC F/M ratio on oxygen transfer in the individual treatment stages. Furthermore, surfactant removal within a two-stage process and the effectiveness of reverse flexing to restore pressure loss of diffusers in the different treatment stages are examined.

P.2.2.1 Energy Efficiency of Two-Stage Activated Sludge Systems

Currently, almost all conventional activated sludge (CAS) wastewater treatment plants operate in an energy-negative mode. When an HRAS system is followed by a second biological treatment stage (e.g., for nitrogen removal), it can be operated differently than a CAS system (Winkler and Widmann, 1994). In this case, the first stage can redirect carbon into waste activated sludge (WAS) through biosorption and energy self-sufficiently remove nutrients (Kroiss and Klager, 2018; Liu et al., 2020). Liu et al. (2020) presented a variety of A-B process designs, and Jimenez et al. (2015) described design parameters to optimize carbon redirection. They defined a typical operation range of HRAS systems as SRT < 1 day, HRT ≈ 30 min, DO < 1 mg $O_2 \cdot L^{-1}$, and very high sludge-specific organic loading rates that result in a concentration of influent particulate, colloidal, and soluble chemical oxygen demand (COD) into the WAS through biosorption. This improves direct energy recovery from carbon-loaded sludge through biogas production (Sancho et al., 2019; Wan et al., 2016). Moreover, in-plant energy consumption is reduced by lower oxygen demand for aerobic carbon removal and higher overall aeration efficiency (Kroiss and Klager, 2018). The separation of carbon- and nitrogen-removing biomass potentially reduces overall oxygen supply by more precise aeration control according to the respective biomass's specific oxygen demand (Svardal and Kroiss, 2011). Depending on the wastewater composition, not enough soluble COD to ensure complete denitrification may be a critical limitation of two-stage processes that is aggravated by additional carbon redirection. Therefore, two-stage WWTPs are recommended for high-carbon or low-nitrogen wastewater treatment; alternatively, they require side-stream short-cut nitrogen removal processes (e.g., nitritation-denitritation or partial nitritation-anammox) to decrease carbon requirement of nitrogen removal (Liu et al., 2020). Nonetheless, two-stage activated sludge configurations are a sustainable option in the ongoing shift from conventional treatment by removal in WWTPs to more energy-efficient treatment and resource recovery in water resource recovery facilities (WRRFs) (Liu et al., 2020).

P.2.2.2 Influences on Oxygen Transfer in Two-Stage Activated Sludge Systems

The α-factor is determined as the ratio of volumetric oxygen mass transfer coefficient in process water compared to clean water as described in ASCE 18-18 (ASCE, 2018) and DWA-M 209 (DWA, 2007). The use of a separate fouling factor (F or αF) to distinguish between diffuser- and wastewater-specific effects on oxygen transfer is described by EPA (United States Environmental Protection Agency, 1989). Recent review articles summarize the influences on oxygen transfer in process conditions. Baquero-Rodríguez et al. (2018) reviewed a variety of factors including diffuser aging and fouling, influent wastewater variability, and airflow rates for fine-pore diffuser aeration. Amaral et al. (2019) focused on the modeling aspect of the gas-liquid transfer in activated sludge. Both studies concluded that the development of a model to consider all factors affecting oxygen transfer in activated sludge systems would be extremely valuable. So far, the complexity of interactions between factors complicates the development of a comprehensive α-model. To achieve this goal, more knowledge about the involved processes has to be acquired.

Therefore, one path to gain deeper insight is to look at extreme variations of activated sludge process designs such as two-stage configurations. The biosorption mechanism utilized for carbon redirection in HRAS stages describes surface adsorption of particulate and colloidal organic matter on sludge flocs and storage of soluble COD inside of biomass (Guellil et al., 2001; Majone et al., 1999). This can have a positive effect on oxygen transfer as substances inhibiting gas-transfer at the bubble-bulk interface are removed or adsorbed on sludge flocs. Garrido-Baserba et al. (2020) discussed strategies to increase oxygen transfer efficiency through biosorption, inter alia by specifically removing surfactants. The amphiphilic structure of surfactants causes a negative effect on oxygen transfer at low concentrations in clean water (Wagner and Pöpel, 1996a) and activated sludge (Rosso et al., 2006). High biosorption of surfactants in a first treatment stage could improve oxygen transfer in a subsequent treatment stage. The overall energy efficiency of an aeration system is determined not only by oxygen transfer in the bulk liquid, but also by pressure loss of diffuser elements. This pressure loss resembles the extra resistance that blowers have to overcome to widen membranes and diffuse air through the membrane perforation. Pressure loss increases due to fouling, aging, and scaling of membranes, and it also negatively affects oxygen transfer efficiency (Garrido-Baserba et al., 2016; Rosso and Stenstrom, 2006). Reverse flexing is a mechanical cleaning method where diffuser membranes are relaxed by turning off the blowers and releasing pressure from the air pipes. This causes a rapid collapse of the diffuser membrane onto the diffuser's frame under hydrostatic pressure.

Turning on the blowers flexes the diffuser's membrane and reopens its slits, which removes biofilm and particulate matter from the membrane surface. As a result, previously built-up pressure loss is mitigated which enables more energy-efficient operation of the aeration system (Odize et al., 2017; Rosso, 2015).

P.2.2.3 Goals of this Study

Factors relevant for energy-efficient operation of aeration systems have been studied for CAS systems; however, comprehensive research is not available for two-stage AS processes. This paper addresses this research gap and defines α-factors for design load cases applicable to design aeration systems of two-stage AS systems by measuring oxygen transfer on a pilot scale. Most importantly, the underlying measurements include variations of diurnal cycle of WWTP operation and influent characteristics, rain and dry weather, and seasonal variations affecting oxygen transfer and the α-factor. The resultant dataset covers various load cases of a two-stage WWTP. Some procedures to design aeration systems use static α-factors, whereas the approach of German guideline DWA-M 229-1 (DWA, 2017), as described in Wagner and Stenstrom (2014), distinguishes three load cases with α_{mean}, α_{min}, and α_{max} factors that we determined accordingly. We also quantified surfactant concentrations in samples throughout the treatment process to examine the distribution of surfactants in the treatment stages of a two-stage configuration. Additionally, different operation of treatment stages within a two-stage system affects bioflocculation capability and resultant sludge composition, which could have an effect on diffuser fouling. We investigated operation and maintenance of fine-bubble diffusers in those conditions through a series of diffuser pressure loss measurements after reverse flexing to determine if fouling can be mitigated effectively in two-stage processes.

P.2.3 Materials and Methods

P.2.3.1 Design and Operation of Pilot-Scale Test Reactors

Long-term ex situ steady-state off-gas monitoring was conducted in pilot-scale test reactors as described in ASCE/EWRI 18-18 (2018). Tank dimensions were 1.2 m × 1.2 m × 5.8 m (L × W × H) with a volume of 8.3 m^3. Two reactors were operated to examine both AS stages of a two-stage process in parallel. Both reactors were equipped with fine-bubble disc diffusers (ELASTOX-T EPDM TYP B, WILO GVA, Wülfrath, Germany) with a diffuser density of 13.5 %. Unlike off-gas measurements using off-gas hoods, the airflow rate within an ex situ reactor can be varied independently from the operation of the WWTP it receives its sludge from. A range of airflow rates (specified for aerated tank volume—$q_{Vol,aer}$) between 0.75 and 2.25 $Nm^3 \cdot m^{-3} \cdot h^{-1}$ was set, covering typical ranges of two-stage WWTPs. Sludge transfer pumps (AGNM02 NEMO®, NETZSCH Holding, Selb, Germany) were operated to maintain a constant hydraulic retention time (HRT) of 15 min as recommended by ASCE/EWRI 18-18. Sludge flow was measured with electromagnetic flowmeters (Promag W 400, Endress + Hauser AG, Reinach, Switzerland). Mixing conditions within the tanks can be assumed close to an ideal continuous stirred tank reactor (CSTR), because of the combined energy input of aeration and sludge transfer.

Mean values of clean water tests of standard oxygen transfer rate (SOTR) were used as a denominator for the α-factor. Clean water tests were conducted with electrochemical dissolved oxygen (DO) probes (Oxymax COS51D, Endress + Hauser AG, Reinach, Switzerland) with a fast response time t_{90} of 30 s. Slower optical DO probes Oxymax COS61D, Endress + Hauser AG, Reinach, Switzerland) were used in process conditions, as long-term testing did not require a fast response time, and their lower maintenance offers more reliable DO measurement in activated sludge operation. While off-gas measurements require a steady inflow, clean water tests were conducted without continuous inflow. In our pilot plant lateral sludge inflow improved oxygen transfer at low airflow rates, which resulted in overestimates of the α-factor. As a consequence, only airflow rates above 0.75 $Nm^3 \cdot m^{-3} \cdot h^{-1}$ were considered in this study. Clean water tests were conducted before and after a long-term off-gas measurement period to evaluate diffuser conditions. This revealed a decrease in SOTR of 2 – 6 % depending on airflow rate and a dynamic wet pressure increase of about 1 kPa. These results primarily indicate inevitable aging of diffusers and secondarily indicate scaling and fouling. Overall, the effect of scaling and fouling during the long-term off-gas measurement was kept low due to monthly pressure cleaning and reverse flexing of disc diffusers twice a week. Therefore, in this study, the oxygen transfer is reported as an α-factor instead of an αF-factor. Additionally, potential biofilm build-up on the reactor tank walls was prevented with monthly cleaning and visual inspection to ensure only suspended biomass transferred from the adjacent full-scale AS tanks was examined in the ex situ reactors for off-gas measurements. Online sensors were cleaned twice a week to prohibit solids deposition and biofilm growth affecting optical instruments.

Other parameters and their sensors and instruments for off-gas measurements were airflow rate measured with thermal mass flowmeters (Proline t-mass A 150, Endress + Hauser AG, Reinach, Switzerland), off-gas concentrations of oxygen (paramagnetic sensor) and carbon dioxide (NDIR) measured with a gas-analyzer (X-STREAM Enhanced, Emerson Electric Co., MO, USA) that receives dry off-gas free of particles (CSS-V, M&C TechGroup, Ratingen, Germany), atmospheric pressure (Cerabar PMC21, Endress + Hauser AG, Reinach, Switzerland), atmospheric temperature (Omnigrad T TST434, Endress + Hauser AG, Reinach , Switzerland), and electrical conductivity (Indumax CLS50D, Endress + Hauser AG, Reinach, Switzerland). Data were recorded in 30 s intervals by online sensors and summarized as 15 min averages. This resulted in high-resolution data that matched the HRT of the test reactors and the interval of operating data provided by the WWTP operator. However, residence time distribution in an ideal CSTR yields a 63 % replacement of activated sludge in the reactors at HRT of 15 min and 98 % at 1 h, respectively. Therefore, for final analysis, 1 h intervals were composed to prevent autocorrelating observations. In total, α-factors were recorded for 9 months in long-term off-gas measurements covering a period of 13 months.

P.2.3.2 Design and Operation of Examined Two-Stage WWTP

The examined two-stage activated sludge WWTP has a design capacity of more than 1 Mio PE. It has a mean dry weather influent flow of 2.6 $m^3 \cdot s^{-1}$ and a maximum wet weather influent flow of almost 7 $m^3 \cdot s^{-1}$ of mostly municipal wastewater, complying with German effluent standards. Raw wastewater is first treated in screens (width 10 mm) and an aerated grit chamber before it flows into the primary clarifier with a mean HRT of 60 min that ranges from 35 to 100 min depending on influent flow. Biological wastewater treatment is split into a first high-rate activated sludge stage for carbon removal and a subsequent second stage for nitrification with a fivefold larger tank volume. Both aerated stages are plug flow reactors with tapered aeration, while 25 % of tank volume of the second stage is a continuously mixed upstream denitrification zone. Both treatment stages have no internal recirculation and are followed by clarifiers that return activated sludge into the respective stages. A bypass line can pass 0.2 $m^3 \cdot s^{-1}$ of primary effluent into the second stage to redirect organic carbon required for biological nutrient removal in the upstream denitrification. A recirculation line can recirculate 0.5 to 0.55 $m^3 \cdot s^{-1}$ of nitrate containing final clarifier effluent into the first stage. This relieves the final downstream denitrification (biofiltration) which removes remaining nitrate. These concepts are described in more detail in Jimenez et al. (2015) and Wandl et al. (2006).

Influent wastewater load is diluted in activated sludge tanks and the concentration of removable substances changes within AS tank zones during treatment, especially in plug flow reactors. Therefore, determining the α-factor of a whole plug flow reactor tank at a certain time requires off-gas testing across all subsequent aeration zones (Redmon et al., 1983; Rosso et al., 2005). However, to closely monitor the diurnal cycle of oxygen transfer, activated sludge was transferred from the front aeration zone of both plug flow aerated stages into the pilot-scale test reactors. Operating data of the first and second stage of the examined two-stage activated sludge WWTP are summarized in Table P.2.1.

Table P.2.1 Operating data of examined two-stage WWTP

Parameter (Abbreviation)	Unit	First Stage		Second Stage	
		Mean ± Std. Dev.	5^{th} – 95^{th} Percentile	Mean ± Std. Dev.	5^{th} – 95^{th} Percentile
Volume Specific Airflow Rate ($q_{Vol,aer}$)	$Nm^3 \cdot m^{-3} \cdot h^{-1}$	1.8 ± 0.5	0.9 – 2.3	0.7 ± 0.2	0.5 – 1.0
Dissolved Oxygen (DO)	$mg \cdot L^{-1}$	0.6 ± 0.3	0.2 – 1.0	3.2 ± 0.2	3.0 – 3.4
Actual Hydraulic Retention Time (HRT_a)	h	0.7 ± 0.1	0.5 – 0.9	1.9 ± 0.3	1.4 – 2.4
Nominal Hydraulic Retention Time (HRT_n)	h	2.0 ± 1.0	0.9 – 3.6	6.2 ± 2.6	2.8 – 10.5
Sludge Retention Time (SRT)	d	1.9 ± 0.7	0.7 – 3.2	31[1] ± 7.3	21 – 44
Total Solids in AS (TS)	$g \cdot kg^{-1}$	3.0 ± 0.4	2.4 – 3.7	6.1 ± 0.6	5.1 – 7.1
Volatile Fraction in AS (MLVSS/MLSS)	%	72 ± 6	63 – 85	59 ± 4	53 – 65
TOC Inflow Concentration (TOC_{in})	$mg \cdot L^{-1}$	75 ± 22	44 – 113	18 ± 5.4	12 – 25
Water Temperature (T_W)	°C	17 ± 3	13 – 22	17 ± 3	13 – 22
Total Suspended Solids in Effluent ($TSS_{effluent}$)	$mg \cdot L^{-1}$	25 ± 12	12 – 46	4.1 ± 1.7	2.1 – 7.6
Sludge Volume Index (SVI)	$mL \cdot g^{-1}$	99 ± 35	51 – 164	49 ± 5.5	41 – 56

[1] Median value; for all other parameters the median deviates by less than 10 % from the above listed means

The sample standard deviation marks the dispersion from mean values during standard operation of the WWTP, while the 5th and 95th percentiles are stated to describe reasonable minimum and maximum operation conditions that are only exceeded in exceptional cases. Volume specific airflow rate $q_{Vol,aer}$ is specified in relation to aerated basin volume. The reported sludge retention time is temperature-corrected to 15 °C (correction coefficient = 1.072, compare Clara et al. (2005)), and outliers outside 1.5 times the interquartile range above and below Q1 and Q3 quartiles were removed. A rolling mean was calculated of the remaining data spanning 2 days for the first stage and 30 days for the second stage. These chosen timespans resemble the median SRT in the respective stages. Online turbidity sensors (SOLITAX sc, Hach Lange GmbH, Düsseldorf, Germany) measuring mixed liquor suspended solids are calibrated for total solids (TS) and regularly compared with laboratory analysis (according to EN 12880). Mixed liquor suspended solids (MLSS) are not measured regularly. On average, MLSS was 0.8 $g \cdot L^{-1}$ lower than TS. Total organic carbon (TOC) inflow concentration (TOC_{in}) considers all inflows of a treatment stage (e.g., supernatant of return activated sludge and bypass flows) proportional to their respective water flow. This combination is required because effluent TOC of the intermediate clarifier recycled into the first stage with return activated sludge has a share of about 30 % of total TOC inflow in the first stage. TOC concentrations are measured by ex situ online analyzers (QuickTOC, LAR, Berlin, Germany) in the influent and effluent of the first stage and drift-corrected to match laboratory analysis (EN 1484). We used TOC as a suitable sum parameter to describe influent wastewater characteristics instead of COD, because ex situ online analyzers of TOC are common in larger WWTPs and enable an analysis with higher temporal resolution than COD laboratory analysis. For comparison, TOC/COD ratios based on laboratory analysis were 0.33 ± 0.05 in the influent of the first stage and 0.46 ± 0.10 in the influent of the second stage. $TSS_{effluent}$ is recorded in the supernatant of the respective clarifier (2 µm pore size). Hydraulic retention time (HRT) refers to the retention time in activated sludge tanks, not the whole treatment stage with clarifiers. It is stated either as nominal HRT_n which considers only influent flow or as actual HRT_a, which includes recirculation flows, as well as main wastewater inflow (compare nomenclature in Henze et al. (2008)). The TOC F/M (feed to mass) ratio is typically derived from TOC concentration in the inflow, MLSS in the AS, and volume of biological treatment stage. To account for dilution in the AS tank and return TOC load of recirculation flows, we use the volume proportional TOC_{in} and HRT_a as described above. To simplify comparison, TS is assumed as given in units of $g \cdot L^{-1}$ similar to MLSS. Thus, we derived an actual TOC F/M_a ratio from parameters given in Table P.2.1 as follows:

$$\text{TOC F/M}_a\text{ ratio} = \text{TOC}_{in} \cdot \text{TS}^{-1} \cdot \text{HRT}_a^{-1} \quad (\text{in kg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}) \qquad \text{(P.2.1)}$$

The use of actual HRT_a, volume proportional TOC_{in}, and resultant TOC F/M_a ratio reflects organic load in the AS tanks more reasonably regarding their effect on oxygen transfer in the front aerated zones than the nominal HRT_n and TOC F/M ratio.

P.2.3.3 Separate Rain and Dry Weather Conditions

This study distinguished rain and dry weather conditions to examine their impact on oxygen transfer in the AS tanks. WWTP operators typically record all-day weather conditions; however, these do not reflect the diurnal inflow dynamic. Instead, we assigned a weather category on the basis of the diurnal variations of collected inflow data. Figure P.2.1 shows the inflow course during diurnal cycle as smoothed functions of percentiles of the inflow represented by the lines. Top and bottom lines describe the percentiles at 0 % and 100 %, while the lines in between depict percentiles from 5 % to 95 % in 10 % steps. The dashed line serves as a distinction where data above were assigned as rain and data below were assigned as dry weather category. It represents the 80th percentile of inflow data based on recorded weather conditions. In the operating data of the examined two-stage WWTP, 77 % of days were recorded as dry weather (dry and frost conditions), while the remaining 23 % were recorded as rain weather (e.g., rainfall, snowfall, and discharge from stormwater retention basins). Therefore, the 80th percentile was chosen to clearly separate rainfall periods from regular operation. A wastewater inflow of 3 $m^3 \cdot s^{-1}$ is considered as rain weather at 6:00 and as dry weather at 12:00. The 85th and 95th percentiles are categorized as rain weather but have a distinct diurnal inflow pattern. While a single rainfall runoff does not follow this pattern, on average, light rainfall is added on the dry weather pattern. In contrast, the 100th percentile represents maximum inflow capacity of the WWTP and is constant throughout the diurnal cycle.

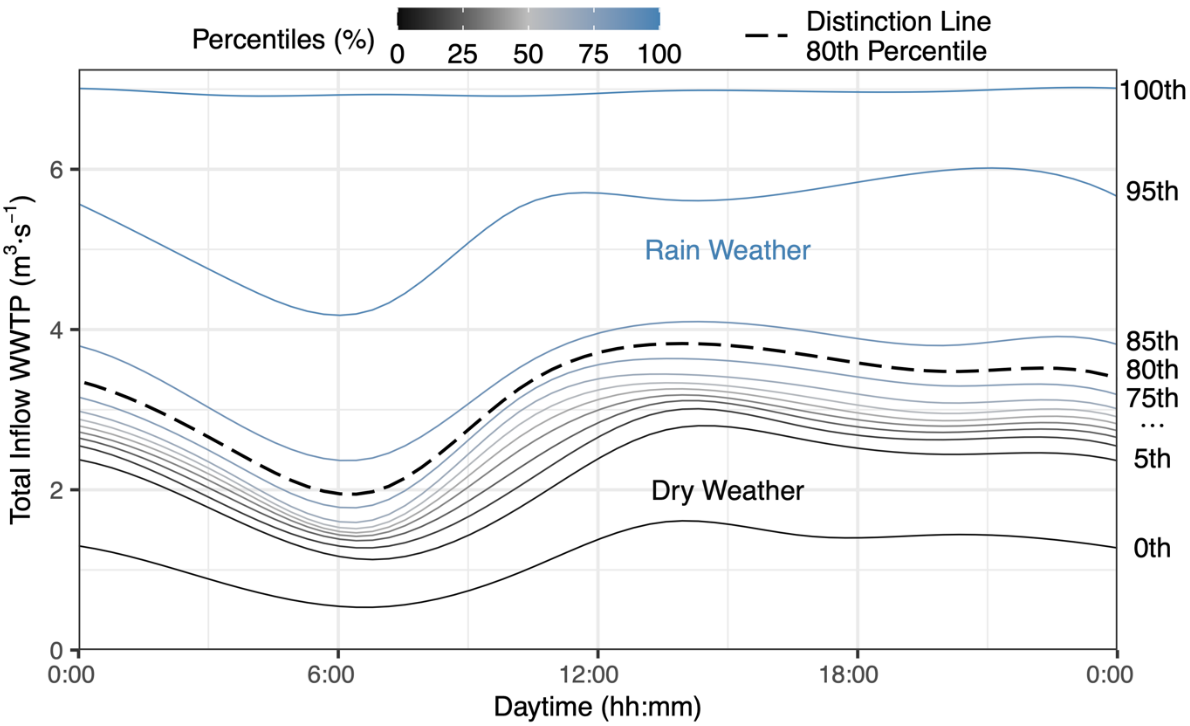

Figure P.2.1 Assigning weather category on the basis of diurnal variations of total wastewater inflow

P.2.3.4 Surfactant Analysis

Surfactant concentrations of successive treatment stages of the two-stage AS WWTP were measured with Hach cuvette tests (Hach Lange GmbH, Düsseldorf, Germany) for anionic (LCK 332), nonionic (LCK 333), and cationic surfactants (LCK 331) in a spectrophotometer (DR 3900, Hach Lange GmbH, Düsseldorf, Germany). Accordingly, 24 h composite samples were taken from primary clarifier influent, first-stage influent, second-stage influent, and second-stage effluent. Grab samples were taken from the first- and second-stage activated sludge tank and settled before taking an aliquot from the supernatant to analyze. The samples were not centrifuged or filtered. However, when taking an aliquot, intake of particles was avoided. Overall, surfactant cuvette tests are error-prone because other surfactant types may cause low-bias results according to working procedure information by the manufacturer. Duplicate measurements of each sample with a recovery were conducted according to the manufacturer's working procedure. The measurement series was repeated three times over the course of 1 year. In total, at least five evaluable tests per surfactant type are available for each sample location with a recovery between 80 % and 120 %.

P.2.3.5 Dynamic Wet Pressure Measurement and Reverse Flexing Procedure

Dynamic wet pressure (DWP), also known as pressure drop, pressure loss, or diffuser headloss, is the pressure difference of a submerged diffuser calculated as the difference between pressure in the air pipe close to the diffuser and the hydrostatic pressure. DWP increases with higher airflow rates; therefore, it is usually specified at a specific airflow rate. Pressure was measured with a capacitive digital pressure transmitter in the air pipes close to the diffuser frame (Cerabar PMC21, Endress + Hauser AG, Reinach, Switzerland). DWP was calculated as the difference of this sensor reading and the hydrostatic pressure in the reactor defined by blow-in water depth, which is limited by an overflow in the test reactors.

Reverse flexing was performed twice a week during maintenance of the pilot reactors, which resulted in a period of 3 to 4 days since the last procedure. To perform reverse flexing, blowers were shut off for up to 2 h and relative pressure in the air pipes was reduced to 0 kPa. The diffusers remained sealed during the long-term measurements as no water leakages were detected in the diffuser frame. Because DWP increases with airflow rate, long-term measurement series were conducted at a constant airflow rate for better comparison. Activated sludge from the first stage was aerated at constant airflow rate of 1.5 and 1.9 $Nm^3 \cdot m^{-3} \cdot h^{-1}$, and sludge from the second stage was aerated at constant airflow rate of 0.8 and 1.0 $Nm^3 \cdot m^{-3} \cdot h^{-1}$. Tests at lower airflow rates were run for 36 days and those at higher airflow rates were run for 26 days. Diffusers were cleaned with high pressure before each measurement series.

P.2.4 Results and Discussion

P.2.4.1 Effect of Rainfall and Diurnal Cycle on Oxygen Transfer

The oxygen transfer in the AS process is subject to a multitude of influence factors that vary seasonally and within daily cycles. Additionally, hydraulic and organic loading differ tremendously between rain and dry conditions, thus affecting oxygen transfer in the activated sludge tanks. Table P.2.2 presents all α-factors measured within this study as described in Section P.2.3 for mean ± standard deviation and 5th and 95th percentiles.

Table P.2.2 α-factors determined with ex situ off-gas measurements in a two-stage WWTP

Parameter (Abbreviation)	Unit	First Stage		Second Stage	
		Mean ± SD	5th – 95th Percentile	Mean ± SD	5th – 95th Percentile
α-Factor (ex situ measurement)	-	0.43 ± 0.06	0.33–0.54	0.80 ± 0.07	0.69–0.91

Figure P.2.2A divides α-factors by treatment stage and weather conditions in an empirical cumulative distribution. The horizontal dashed lines mark the 5th and 95th percentiles. Lower mean α-factors were measured in the first stage (0.43) than in the second stage (0.80), as indicated by the vertical dashed lines. Kroiss and Klager (2018) stated similar α-factors of 0.45 and 0.7 in first and second stages of the Vienna main wastewater treatment plant. Overall, influences affecting oxygen transfer differ tremendously between the first and second stage in a two-stage AS configuration. In particular, the first high-rate stage cannot be compared with CAS systems, where α-factors for systems with nitrification and denitrification typically fall into the range of 0.6 to 0.75 (Rosso et al., 2008a). Additionally, the distinction of rain and dry weather reveals that α-factors in the first stage decreased during high inflows of rainwater, whereas no such effect was apparent in the second stage. The effect of stormwater runoff on oxygen transfer has not been discussed in the literature so far. However, rain events have an impact on multiple parameters potentially affecting oxygen transfer in the activated sludge tank, as shown before. Stormwater runoff affects the hydraulic and influent load of a WWTP. A first flush often brings a high load due to washout of sewer sediments followed by slightly contaminated rainwater afterward (Larsen et al., 1998). Wilén et al. (2006) concluded that biological processes in the sewer system are more aerobic at high flows and more anaerobic at low flows, thus changing wastewater properties. Typical effects of rain events also include lower conductivity and water temperature with increased total inflow (data not shown), which is compensated for by standardization to norm conditions when determining α-factors.

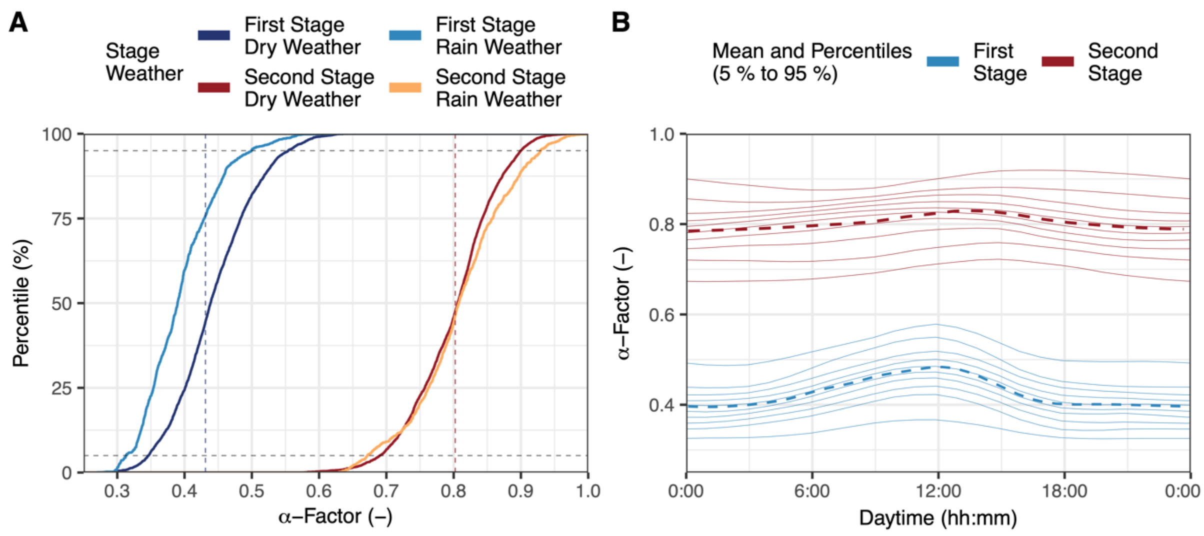

Figure P.2.2 Empirical cumulative distribution (A) and diurnal variation (B) of α-factors as percentiles (solid lines) and means (dashed lines) in the examined two-stage AS WWTP

Figure P.2.2B shows the diurnal variation of recorded α-factors in both stages. The lines represent the course of percentiles from 5% to 95% as described in Section P.2.3.3 for Figure P.2.1. The first stage was characterized by a distinct peak of the α-factor at noon, regularly fluctuating between 0.39 and 0.48, as indicated by the dashed line representing the mean α-factor. Peak α-factors are measured during daytime instead of nighttime due to a long retention time of wastewater in a large sewer system. In contrast, α-factors in the second stage had a smoother course without a distinct peak. Here, α-factors fluctuated on average between 0.78 and 0.83 within a day. The influent load into the second stage was decreased and buffered by the preceding HRAS tank and upstream denitrification zone, resulting in a smoother diurnal cycle of α-factors. This also explains the different extent of rain effects on oxygen transfer in two-stage AS treatment stages, as further discussed below.

The diurnal cycle of α-factor observed in the first stage was previously described by an inverse relationship of α-factor and influent load (Jiang et al., 2017; Leu et al., 2009). For operators of WWTPs, this negative correlation means that oxygen transfer is generally at its lowest when oxygen demand is highest. To illustrate this relationship, Figure P.2.3 displays the volume specific airflow rate ($q_{Vol,aer}$) in the full-scale AS tanks as the dependent variable of TOC inflow concentrations (TOC_{in}) and α-factor. Blowers were controlled by DO in the aeration basins to set the airflow rate. First, Figure P.2.3A shows that volume specific airflow rate was increased in response to higher TOC_{in} to meet resulting oxygen demand of biomass in both stages. Secondly, lower α-factors forced operators to increase airflow rates to meet this oxygen demand, as shown in Figure P.2.3B. This figure also reveals that this relationship was more distinct in the first stage than the second. The two stages also differed during rain weather, where lower α-factors coincided with higher airflow rates in the first stage, but no significant decrease in α-factor was apparent in the second stage. It is important to note that α-factor is usually not affected by airflow rate directly, but rather coincides with changes in oxygen demand due to influent load (Gillot and Héduit, 2000; Rosso et al., 2005).

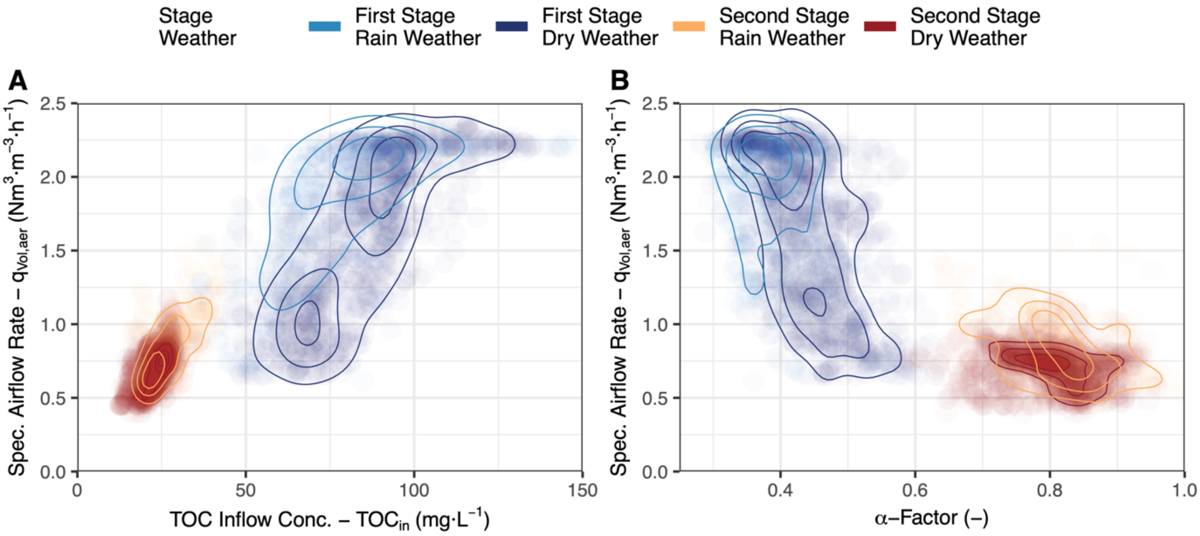

Figure P.2.3 Volume specific airflow rate of full-scale aeration basins for TOC_{in} (A) and α-factor (B) grouped for treatment stages and weather conditions

In Figure P.2.3 the individual points represent mean data recorded within 1 h intervals. Colors distinguish between rain and dry weather periods as specified in Section P.2.3. To visualize the two-dimensional distribution of the resulting clusters, they were divided by three density lines with each interval containing 25 % of the respective cluster data. A smaller area enclosed within these density lines denoted a higher density of the contained data points.

Overall, these results show that oxygen transfer in the second stage was more stable than in the first stage. It is important to emphasize the resultant effect on the required airflow rate to meet oxygen demand in the treatment stages; the described daily fluctuation of α-factor from 0.48 to 0.39 in the first stage required an increase of 22 % of the airflow rate to compensate for oxygen transfer inhibition. In comparison, a decrease from 0.83 to 0.78 in the second stage required adjustment of airflow rates of only 6 % within a typical day. Moreover, Table P.2.2 and Figure P.2.2 reveal the range and distribution of potential α-factors in the two stages caused by various influences on oxygen transfer.

P.2.4.2 Influence of Organic Loading on Oxygen Transfer

Below, we further examine influences that resulted in the presented range of α-factors. The TOC F/M ratio is a suitable aggregate parameter that correlates with oxygen transfer inhibition (Günkel-Lange, 2013). Figure P.2.4 displays four scatterplots of measured α-factors for TOC F/M_a ratio and its individual components: actual hydraulic retention time (HRT_a), TOC inflow concentration (TOC_{in}), and total solids (TS).

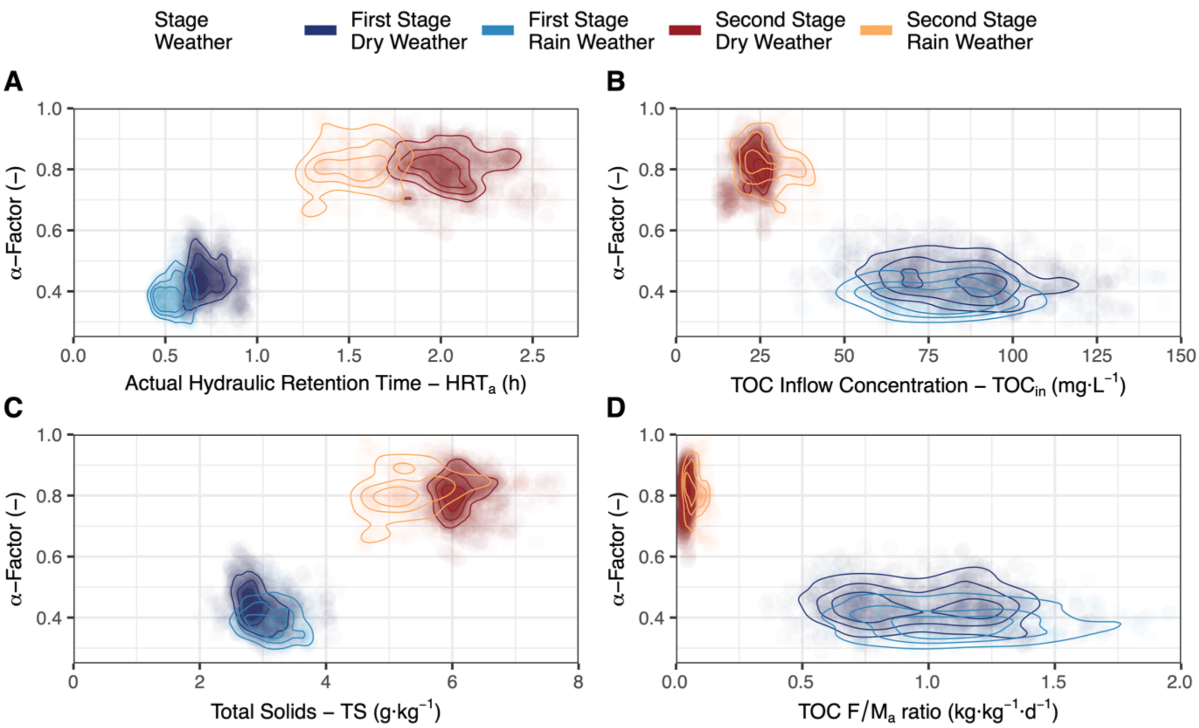

Figure P.2.4 The α-factors for HRT_a (A), TOC_{in} (B) TS (C), and the aggregated parameter TOC F/M_a ratio (D) grouped for treatment stages of a two-stage WWTP and weather conditions

Figure P.2.4A shows the α-factors recorded in the first and second treatment stages at their respective HRT_a. The treatment stages of the examined two-stage WWTP were operated differently and, as a result, all diagrams in Figure P.2.4 clearly distinguish both stages from each other. Moreover, rain and dry weather categories were clearly separated within treatment stages, as HRT_a reflects high and low water inflow. Overall, lower α-factors were recorded in the first treatment stage with its shorter HRT_a. The longer HRT_a within the first stage indicated slightly higher α-factors, while no such effect could be seen in the second stage. Although water inflow and the resultant HRT_a have no known direct impact on oxygen transfer, a change of hydraulics in a WWTP affects other parameters that have an impact on the α-factor.

TOC inflow concentrations in the first stage were higher and spread over a wider range than in the second stage, as displayed in Figure P.2.4B. Roughly two-thirds of TOC influent load was removed in the first stage. While Figure P.2.3 suggests a clear correlation between α-factor and TOC influent concentration, Figure P.2.4B shows that it was less evident within the respective treatment stages. However, looking at both treatment stages, a negative correlation between TOC inflow concentration and oxygen transfer can still be confirmed. Jiang et al. (2017) concluded a similar negative logarithmic relationship between α and COD on the basis of measurements in three WWTPs. Ahmed et al. (2021) applied a power function to fit an α-model for SBR reactors. Both approaches came to similar results to this study but examined different

WWTP process configurations that are not directly comparable to the examined two-stage process. The major difference between α-factors in treatment stages can be attributed to the oxygen transfer inhibiting characteristics of readily biodegradable substrate (Ahmed et al., 2021), especially accumulation of surfactants on the bubble surface (Rosso et al., 2008a; Wagner and Pöpel, 1996b). During rain periods, α-factors observed in the first treatment stage were lower than in dry conditions, although TOC inflow concentrations were similar or lower. However, TOC load increased when considering the increased water flow and organic load of a first flush in the sewer system as a result of a rainfall event, thus explaining lower α-factors. This effect was not apparent in the second stage. Here, TOC inflow concentration was slightly higher during rainy weather as some organic load remained untreated at low HRT_a in the first stage. Nonetheless, α-factors in the second stage did not decrease because most influent organic load was buffered in the first stage and the upstream denitrification zone of the second stage. The implementation of an upstream denitrification stage has been reported as advantageous for oxygen transfer in CAS systems (Rosso and Stenstrom, 2005). Thus, the high α-factors in the second stage can be attributed in part to this, even though some readily biodegradable substrate was passed into the second stage by the bypass line.

Figure P.2.4C displays α-factors for total solids (TS). The α-factors and TS in the second stage were higher than in the first stage and high for activated sludge process in general. Within the second stage, no correlation with TS was indicated, while a slight decrease in α was apparent in the first stage, coinciding with rain weather. This outcome is discussed in more detail below.

TOC F/M_a ratio in Figure P.2.4D combines the previously discussed parameters. Its course was similar to TOC_{in} in Figure P.2.4B except for the first stage during rainfall events. Here, high water inflow and TOC concentration produced higher TOC F/M_a ratios with a negative effect on α-factor. Günkel-Lange (2013) examined the relationship between COD F/M ratio and α-factor for extended aeration, nitrogen-removal, and carbon-removal CAS systems and proposed an inverse linear correlation. Again, the examined two-stage WWTP is different from CAS systems and complicates direct comparison. However, the presented data complement the understanding of oxygen transfer dynamics in more complex WWTP process configurations.

According to the diagrams in Figure P.2.4, oxygen transfer in the second treatment stage was seemingly unaffected by any variation of the presented parameters. However, this cannot be concluded from the above analysis with certainty, as at most only two interactions were taken into account in each diagram. Furthermore, the combined parameter TOC F/M_a ratio obscured variation of its individual components (e.g., 100 kg/h TOC load at 3 g/L TS would result in the same TOC F/M ratio as 200 kg/h TOC load at 6 g/L TS, but the resulting conditions would affect oxygen transfer differently). Considering both treatment stages, our results confirm the inverse relationship between TOC_{in} or TOC F/M_a ratio and α-factor, as presented in previous studies. However, no single parameter illustrated in Figure P.2.4 correlated significantly with the α-factor when considering oxygen transfer in individual treatment stages.

P.2.4.3 Interaction of Suspended Solids and Hydraulic Load with Oxygen Transfer

Generally, TSS concentration, usually measured as mixed liquor suspended solids (MLSS), inversely correlates with the α-factor. This has been extensively demonstrated for membrane bioreactors (MBR), where different rheology of thick sludge at MLSS up to 30 $g{\cdot}L^{-1}$ has an influence on gas transfer dynamics (Cornel et al., 2003; Germain et al., 2007; Krampe and Krauth, 2003). Henkel (2010) proposed that the volatile fraction of suspended solids (mixed liquor volatile suspended solids—MLVSS) in particular causes oxygen transfer inhibition. These studies extrapolated the inverse relationship measured in MBRs into conventional activated sludge systems (CAS), where typical MLSS concentrations are below 6 $g{\cdot}L^{-1}$. In contrast, newer studies stated that biosorption decreases the concentration of organic substances in the soluble phase, thereby reducing oxygen transfer and inhibiting accumulation in the gaseous phase (Ahmed et al., 2021; Odize, 2018). Higher MLSS increases the biosorption of organic matter in CAS, which additionally improves carbon redirection in HRAS stages (Jimenez et al., 2015; Rahman et al., 2016). As a consequence of biosorption as the dominant impact on oxygen transfer, a positive correlation between MLSS concentrations up to 6 $g{\cdot}L^{-1}$ and α-factor was proposed by Baquero-Rodríguez et al. (2018). Overall, there seems to be no robust relationship between MLSS and α-factor for CAS (Ahmed et al., 2021). Modeling α from MLSS does not include possible influences of floc structure on oxygen transfer, which vary inevitably between WWTPs. It is probable that floc size (e.g., measured as particle size distribution), settling characteristics (SVI), or addition of precipitants (e.g., for phosphorus removal) alter the liquid-solid interface, thus also influencing the gas-liquid and gas-solid interfaces. To summarize, MLSS or TS as typical parameters in wastewater treatment cannot describe all properties of the solid and liquid phase that are relevant to the dynamic of oxygen transfer once the gas phase is added.

Below, we discuss various parameters to describe the solid and liquid phase in the treatment stages of the examined two-stage WWTP and their potential influence on the α-factor. As shown in Figure P.2.3C, total solids were overall higher in the second stage (6.1 ± 0.6 $g{\cdot}kg^{-1}$) than in the first stage (3.0 ± 0.4 $g{\cdot}kg^{-1}$). In contrast, the volatile fraction of the respective sludges was higher in the first stage (72 ± 6 %) than in the second stage (59 ± 4 %). Although Henkel (2010) argued that the inverse relationship between the α-factor and the solid phase is better described by MLVSS than MLSS, this is not immediately obvious when comparing the absolute MLVSS in the two-stage

WWTP. Here, MLVSS was still higher in the second stage (~3.6 $g \cdot L^{-1}$) than in the first stage (~2.2 $g \cdot L^{-1}$), even though α-factors were higher in the second stage. Thus, in our results, a potential negative effect of organic content of sludge measured as MLVSS was superimposed by enhanced biosorption in the second stage, ultimately increasing oxygen transfer. This is supported by various characteristics that could be beneficial to oxygen transfer in the second stage compared to its preceding first stage, such as better sludge settling (SVI of 49 ± 5.5 $mL \cdot g^{-1}$ compared to 99 ± 35 $mL \cdot g^{-1}$). This would also result in lower total suspended solids in effluent (4.1 ± 1.7 $mg \cdot L^{-1}$ in second stage instead of 25 ± 12 $mg \cdot L^{-1}$ in first stage). The activated sludge was also altered by addition of sodium aluminate as precipitant for phosphorus removal in the influent and effluent of the second stage. Overall, this also affected the liquid phase, which had a visually distinguishable higher turbidity of supernatant from the first-stage activated sludge compared to the clear supernatant of sludge samples from the second stage. SVI, $TSS_{effluent}$, precipitant use, or turbidity of supernatant have not previously been used to explain oxygen transfer in the AS process. Their individual influence on oxygen transfer cannot be quantified, because only two stages with opposed characteristics were examined in our study. However, these parameters further describe characteristics of the solid phase within the two-stage process that could explain the overall difference of α-factors between the first and second stage.

Within the second stage, no correlation of α-factor with TS was indicated, whereas a slight decrease in α was apparent in the first stage, coinciding with rain weather, as depicted in Figure P.2.3C. Rainfall affected TS concentrations differently in the treatment stages of the two-stage WWTP. Figure P.2.5A illustrates the relationship between TS and HRT_a for both rain and dry weather inflow in the respective treatment stage. At lower HRT_a and high hydraulic load during rainy weather, TS decreased in the second stage, while it remained stable in the first stage. This is unexpected as processes with higher HRT and SRT are generally less susceptible to biomass washout due to stormwater flows (McMahan, 2006; Tchobanoglous et al., 2014). Examining operating data indicated that this may have been caused by washout of TS from the primary clarifier into the first stage at shorter HRT_a (data not shown). However, the elevated TS concentrations might not have been the only cause of lower α-factors during stormwater treatment in the first stage. HRT_a represents the possible adsorption contact time of soluble and colloidal organic substances with sludge flocs within the AS tank. Once this organic load is adsorbed on sludge flocs, it is removed through waste activated sludge in the clarifier, and it is also less likely to inhibit oxygen transfer in the gas phase. Jimenez et al. (2015) determined optimal operating conditions of an HRAS system (260 L, CSTR) for removal of soluble, colloidal, and particulate COD at HRTs of >15 min, >30 min, and >45 min, respectively. As a conclusion, low HRT_a caused by rainwater inflow decreased biosorption capacity in the first stage which left more soluble and colloidal organic substances that could accumulate in the gas phase, thus decreasing the α-factor. On the contrary, the α-factor did not drop at lower HRT_a and TS in the second stage (see Figure P.2.3A,C). However, as the second stage received low organic load (see Figure P.2.3B), biosorption mechanisms most probably were much less pronounced than in the first stage.

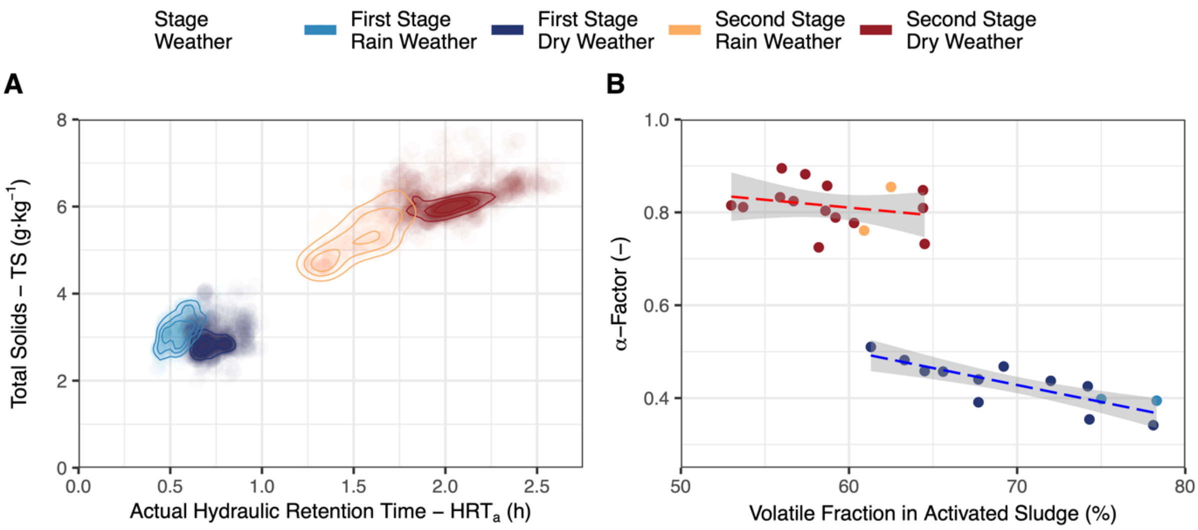

Figure P.2.5 TS for HRT_a (A) and α-factor for volatile fraction in activated sludge as daily mean (B), grouped for both treatment stages of a two-stage WWTP and weather conditions

The α-factors are summarized as daily mean values in Figure P.2.5B and compared with volatile fraction determined from grab samples of activated sludge. Overall, the volatile fraction was lower in the second stage than the first stage due to lower load, higher SRT, and the addition of sodium aluminate as precipitant for phosphorus removal. Within the treatment stages, the regression line surrounded by 95 % confidence intervals revealed a negative correlation of α-factor with volatile fraction in the first stage. While an effect potentially remained, no significant correlation was apparent in the second stage. Operating data revealed a slightly elevated volatile fraction in activated sludge, as well as return activated sludge, at lower HRT_a (data not shown), which could have further decreased α-factor during rainy weather. As a conclusion, the suggested negative correlation of α-factor with the volatile fraction of solids by Henkel (2010) is one of the mechanisms determining oxygen transfer dynamics within the first stage. A stronger impact of volatile fraction was demonstrated in the first stage, whereas, in the second stage, it was superimposed by other influences.

It is worth mentioning that the individual impact of wastewater parameters on α-factor discussed in this study cannot be derived and quantified from the above analysis. In contrast to a controlled experimental design in which all examined parameters are varied systematically, we measured oxygen transfer of an operating full-scale WWTP. The resulting dataset describes only a combination of parameters occurring in real conditions. Additionally, building a mechanistic model of influences on oxygen transfer with a multivariate analysis produces unreliable results when based only on two AS stages that are operated as differently as in the examined two-stage WWTP. The diagrams in Figure P.2.3 show no overlap between α-factors measured in the treatment stages and their process parameters. Hence, complementing our results with further data from CAS systems is necessary to fill these gaps and enable more general inference from wastewater treatment parameters on oxygen transfer dynamics. Lastly, although treatment capacity and overall oxygen demand certainly change throughout seasons, no strong seasonality of α-factor can be derived from our results thus far. Nonetheless, our results allow a complete assessment of α-factors for aeration system design purposes in a two-stage WWTP.

P.2.4.4 Design Load Cases for Aeration Systems of Two-Stage WWTPs

The design of aeration systems of WWTPs specifies the number of diffusers and airflow rates to meet oxygen demand in activated sludge tanks. Diffuser manufacturers state standardized oxygen transfer parameters determined in clean water. However, to consider oxygen transfer inhibition occurring in activated sludge, these parameters have to be multiplied by the α-factor. This design process has been described in various technical guidelines and reference books (DWA, 2019; United States Environmental Protection Agency, 1989; Water Environment Federation, 2018). Oxygen transfer inhibition depends on the WWTP's treatment goal and various processes, among other factors. However, no α-factors have been proposed for two-stage WWTP process configurations thus far. Therefore, according to our results from long-term measurements, we propose α-factors for the design of aeration systems in two-stage systems.

The design approach of German standard DWA-M 229-1 (DWA, 2017), based on Günkel-Lange (2013), applies mean, minimum, and maximum α-factors to define load cases. The α_{mean} represents the average operation conditions of a WWTP. We, therefore, calculated α_{mean} as the average of all α-factors measured during dry weather operation at the examined two-stage WWTP that fell between the mean ± standard deviation of HRT_a, TS, and TOC_{in}, as stated in Table P.2.1. From this, we derived α_{mean} values of 0.45 and 0.80 for the first and second stages, respectively. Because no rainy weather was considered for αmean, it was slightly higher than the average of all measurements in the first stage (0.43), while there was no difference in the second stage (0.80, compare Table P.2.2). The α_{min} and α_{max} values describe oxygen transfer inhibition during high and low load of the WWTP, respectively. We defined these α-factors on the basis of a comprehensive dataset including seasonal variation, as well as rain and dry weather conditions, measured within a 13 month period of conducting long-term off-gas measurements. Hence, we approximated αmin and αmax as the 5th and 95th percentiles of the full dataset, respectively. These percentiles were chosen with a remaining measurement uncertainty in mind. If the design process requires otherwise, the full set of measured data is shown in Figure P.2.2. Our proposed α-factors to design aeration systems in two-stage configurations are summarized in Table P.2.3. These results are applicable for the design of aeration systems in two-stage WWTPs similar to the one examined in this study.

Table P.2.3 The α-factors for design load cases of two-stage activated sludge WWTPs

Treatment Stage	α_{mean} **(-)**	α_{min} **(-)**	α_{max} **(-)**
First Stage (HRAS)	0.45	0.33	0.54
Second Stage	0.80	0.69	0.91

P.2.4.5 Removal of Surfactants in Two-Stage WWTPs

Surfactants have a negative effect on oxygen transfer even at low concentrations due to their amphiphilic structure. They adsorb on the gas-liquid interface of bubbles, as well as on the solid phase of sludge flocs and other particles. Quantifying surfactant loads throughout the wastewater treatment process allows identifying which treatment stage is particularly affected by oxygen transfer inhibition and which treatment process eliminates surfactants. Although a decrease in surfactant concentrations with each treatment stage is expected, the extent of such a reduction is not obvious in two-stage configurations. Effluent quality of a HRAS stage is poor because it is followed by a second treatment stage. First-stage settling tank effluent is characterized by a visible turbidity, remaining mean TOC of 48 mg·L^{-1}, and TSS$_{effluent}$ of 25 mg·L^{-1} (see Table P.2.1). Thus, the remaining surfactant concentration passing into the second stage cannot be neglected for oxygen transfer and has to be measured.

Figure P.2.6 shows boxplots of surfactant concentrations of successive treatment stages of the examined two-stage WWTP divided into three surfactant types. The median of each surfactant type in a sample is summed and connected by a dashed line (median total). Boxplots and the trendline show that surfactant concentrations decreased throughout the treatment stages. Most importantly, total surfactant concentration decreased about 70 % from first-stage influent to second-stage influent, and a dilution of influent concentration in both treatment stages was apparent, as concentrations in the activated sludge supernatant were lower than the preceding influent concentrations. Anionic and nonionic surfactants were more prevalent in the samples, which is typical for municipal wastewater composition (Fraunhofer Institut UMSICHT, 2003; Petrovic and Barceló, 2004). Although absolute concentrations of individual cuvette tests are unreliable, the performed measurement series provides a reasonable span of concentrations for each treatment stage. In comparison, Odize (2018) measured anionic surfactants in HRAS influent (8 ± 2 mg·L^{-1}) and effluent (1 ± 0.1 mg·L^{-1}), both of which are within the above described surfactant concentration range. The overall surfactant removal of more than 95 % within the WWTP is in line with other studies

(Petrovic and Barceló, 2004). The high surfactant concentrations measured in the first treatment stage correspond to low α-factors (0.43 ± 0.06), as well as lower surfactant concentrations and higher α-factors (0.80 ± 0.07) in the second stage. Hence, the previously described higher alpha values in the second stage can also partially be attributed to the adsorption and biological removal of surfactants in the first stage.

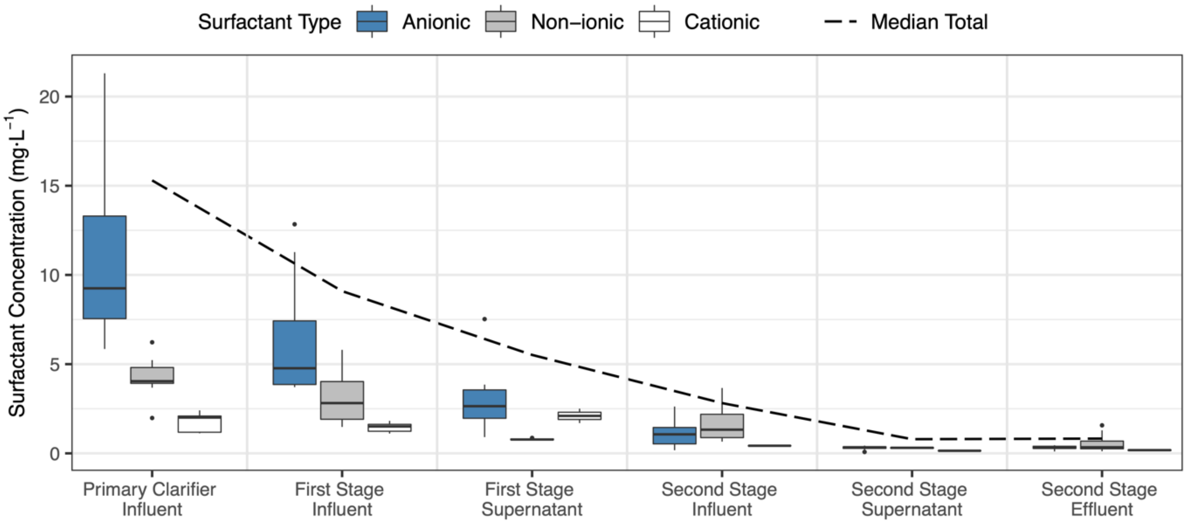

Figure P.2.6 Surfactant concentrations of successive treatment stages divided into surfactant types

P.2.4.6 Reverse Flexing in Two-Stage Processes

Influencing factors on fouling in biological wastewater treatment have been studied extensively for membrane bioreactors (Le-Clech et al., 2006), whereas the effect of fouling on diffuser membranes has focused primarily on quantifying economic implications (Garrido-Baserba et al., 2017, 2016; Rosso et al., 2008b). Knowledge about site-specific wastewater characteristics and WWTP operation on fouling of diffuser membranes is sparse. Thus, Rosso et al. (2012) even suggested implementing on-site long-term column testing of various diffusers as part of the design procedure to take site-specific fouling effects into account when selecting diffusers. As discussed before, inflow wastewater characteristics in the treatment stages of the examined two-stage WWTP and their operation differ; therefore, sludge characteristics differ as well. The resulting separated biomasses with higher content of heterotrophic organisms in the first stage for high-rate carbon removal and autotrophic organisms in the second stage for nitrification could affect fouling behavior of diffusers differently. So far, it is unknown whether existing diffuser maintenance procedures can be applied to mitigate the pressure loss of diffusers in two-stage WWTPs.

Figure P.2.7 shows the boxplots of measured DWP within 12 h intervals after reverse flexing was performed. Median values revealed an expected increase of DWP within the typical 3.5 day interval between maintenance. Most interquartile ranges spanned less than 1 kPa of DWP difference except the test series in the first stage at 1.9 $Nm^3 \cdot m^{-3} \cdot h^{-1}$, where airflow rate fluctuated by ± 0.5 $Nm^3 \cdot m^{-3} \cdot h^{-1}$ due to blower limits. Within the test series, no systematic increase in DWP during multiple cleaning intervals was observed (data not shown), which would be expected over longer periods without periodic pressure cleaning (Rosso, 2015; Rosso and Stenstrom, 2006). According to these test series, we can conclude that pressure loss can be restored effectively with reverse flexing in both treatment stages of a two-stage WWTP. In conclusion, operators of a two-stage WWTP do not have to adapt different diffuser maintenance intervals or procedures for the two treatment stages.

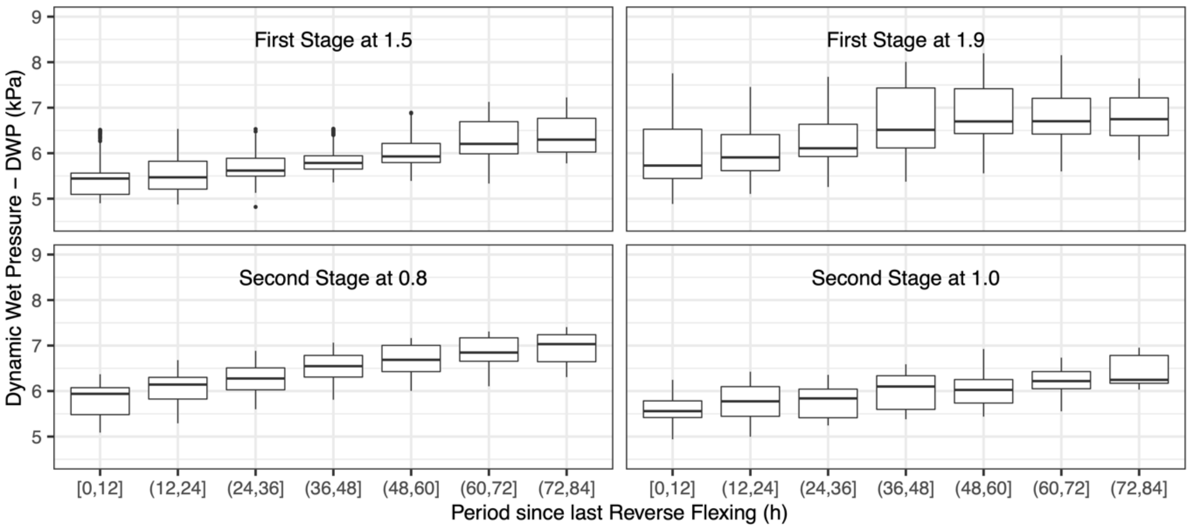

Figure P.2.7 Increase in DWP of disc diffusers since last reverse flexing procedure during operation in activated sludge from first and second stage and at two specific airflow rates ($Nm^3 \cdot m^{-3} \cdot h^{-1}$)

P.2.5 Conclusions

On the basis of our long-term off-gas measurements, we summarize below our findings relevant for design and operation of aeration systems in two-stage activated sludge WWTPs.

- This paper defined α-factors for the first and second stages of a two-stage WWTP. The underlying off-gas measurements on a pilot scale covered a typical range of operation conditions of such a process, as detailed in Table P.2.1, including seasonal variation, as well as dry and wet weather conditions. As a result, α-factors for design load cases were derived for practical application to design aeration systems more accurately. They were determined as 0.45 for α_{mean} and 0.33/0.54 for $\alpha_{min}/\alpha_{max}$ in the first stage (HRAS), and as 0.80 for α_{mean} and 0.69/0.91 for $\alpha_{min}/\alpha_{max}$ in the second stage. Because different process configurations of two-stage processes exist, these α-factors can be transferred to configurations similar to the one examined in this study. No range of α-factors for two-stage processes was previously proposed.

- Our results show how key operating parameters influence the oxygen transfer in the activated sludge system. Most importantly, the impact of high TOC concentrations in inflow resulting in lower oxygen transfer rates can be confirmed and quantified for a two-stage activated sludge process. TS and HRT_a in the treatment stages were affected differently by stormwater treatment. As a result, α-factor decreased in the first stage, whereas the second stage remained unaffected during high wastewater inflow. Hence, engineers can more accurately decide whether an aeration system design meets the demands of a similar WWTP to that examined in this study. Nonetheless, individual wastewater parameters cannot describe α-factor due to various interacting influences. Therefore, applying machine learning methods to predict oxygen transfer is a multivariate approach that we will examine in the future.
- Inflow surfactant concentrations measured in 24 h composite samples revealed that surfactant load was significantly lower in the second stage compared to the first stage. Surfactants had a disproportionate influence on oxygen transfer compared with TOC. Lower α-factors in the first stage could be attributed to this effect but not quantified specifically for surfactants compared to TOC in general.
- The positive effect of reverse flexing as a maintenance method to restore dynamic wet pressure was observed in both stages. There was no significant difference in fouling effect on diffusers, although sludge composition differed tremendously between the high rate and nitrification stage. Therefore, operators of two-stage WWTPs do not have to adapt different maintenance intervals when planning a reverse flexing schedule.

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P.3 Dynamic alpha factor prediction with operating data - a machine learning approach to model oxygen transfer dynamics in activated sludge

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Keywords: aeration, dynamic α-factor, data-driven model, dynamic prediction, random forest, wastewater treatment

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P.3.1 Abstract

Aeration is an energy-intensive process of aerobic biological wastewater treatment. An accurate model of oxygen transfer dynamics in activated sludge tanks would improve design and operation of aeration systems. Such a model should consider spatial and diurnal variation of α-factor as well as site-specific conditions that impact oxygen transfer. For this dynamic prediction a machine learning approach was used for the first time. The data-driven method was based on long-term ex-situ off-gas measurements with pilot-scale reactors (5.8 m height, 8.3 m^3 volume) coupled to full-scale activated sludge tanks on the sites of two conventional and a two-stage activated sludge treatment plant. The ex-situ off-gas method allowed to quantify theoretical off-gas parameters in non-aerated zones and thus consider the whole activated sludge tank. We introduced the α_0-factor to compare aerated and non-aerated zones under nonsteady-state conditions. Like the established α-factor for steady-state conditions, the α_0-factor describes oxygen transfer inhibiting effects in activated sludge. α_0-factor was lowest in upstream denitrification zones. This indicates an anoxic elimination of oxygen transfer inhibiting wastewater contaminants which improved oxygen transfer in subsequent aerobic zones. Random Forest models predicted α_0-factor reliably in all examined activated sludge tanks even for stormwater events and seasonal variation. Model development only required online sensor data already available to operators. Our results suggest that machine learning models can dynamically predict α-factors in a variety of activated sludge processes, thus considering site-specific conditions in model training without manual calibration.

P.3.2 Introduction

The oxygen transfer in activated sludge systems depends on a wastewater treatment plant's (WWTP) process layout as well as wastewater and activated sludge characteristics and can be summarized as a three-phase system. These phases consist of rising air bubbles as the gas phase, wastewater with dissolved substances as the liquid phase and particulate substances, especially activated sludge flocs, as the solid phase. Accurately modeling the oxygen transfer in activated sludge can lead to significant energy savings (Bencsik et al., 2022; Jiang et al., 2017) as aeration is an energy-intensive process in biological wastewater treatment (Reardon, 1995; Rosso et al., 2011). The α-factor is used to summarize oxygen transfer inhibiting effects as the ratio of oxygen transfer in process water and clean water and can be measured with off-gas methods (ASCE 18-18, 2018; DWA-M 209, 2007).

Recent reviews in the research field of aeration technology discussed several influences on oxygen transfer based on the α-factor (Baquero-Rodríguez et al., 2018) and general modeling of mass transfer in activated sludge (Amaral et al., 2019). Research findings from studies conducted over the past decades examined and modeled various influences on oxygen transfer. For example, Wagner and Pöpel (1998) investigated the influence of different submergence depths and diffuser densities on oxygen transfer efficiency in clean water oxygen transfer tests; Gillot et al. (2005) performed a dimensional analysis with parameters that describe the tank geometry; Rosso et al. (2005) modeled the effect of specific airflow rate and sludge retention time (SRT) on α-factors from a database of off-gas measurements collected over 15 years from 30 different activated sludge tanks; Rosso and Stenstrom (2005) reported improved aeration efficiency in conventional activated sludge (CAS) systems with an upstream denitrification stage; Jiang et al. (2017) considered diurnal variations by modelling dynamic α-factors based on COD concentrations; Henkel (2010) developed a model that provides a negative correlation between total suspended solids (TSS) concentration and α-factor for TSS > 6 $g \cdot L^{-1}$, while Baquero-Rodríguez et al. (2018) argued that for TSS < 6 $g \cdot L^{-1}$ a positive correlation with the α-factor exists due to biosorption of oxygen transfer inhibiting substances; Amaral et al. (2017) discussed the necessity to include blower performance, pressure drop of pipes and diffusers, and other tank geometry and controller specific parameters to model gas transfer; and Bencsik et al. (2022) combined several factors into a model (e.g., COD, SRT, TSS, and position in activated sludge tank) to account for spatial and temporal variations of the α-factor.

From a practical point of view, reliable modeling of α-factors would enable more accurate design and more energy-efficient operation of aeration systems than assuming constant α-factors (Ahmed et al., 2021; Bencsik et al., 2022). However, although many parameters have been investigated, no generally valid model to determine the α-factor has been found yet (Amaral et al., 2019). Published results are fragmented and differ regarding essential parameters (e.g., geometry and type of aeration system, aeration tank layout, WWTP process layout, and wastewater characteristics). Consequently, there is still no sufficient data base to develop a mechanistic model of oxygen transfer that can be generally applied to aeration systems of any WWTP.

In recent years, application of online measurement for continuous monitoring and process control of WWTPs has increased tremendously as sensors have become more reliable. Online sensors record primary parameters that describe the mass balance of the activated sludge process (e.g., flow rates, influent, and effluent concentrations of wastewater contaminants) and secondary parameters that describe further process conditions (e.g., dissolved oxygen concentration (DO), pressure, water temperature, electrical conductivity). These parameters are commonly used to describe the biological treatment process in WWTPs as mechanistic, white-box models, e.g., activated sludge models (ASM) (Henze et al., 2000). In contrast, data-driven machine learning (ML) methods produce black-box models where the exact function between an output (dependent or response variable) and inputs (predictor variables or features) is unknown. A dataset in supervised machine learning consists of pairs of predictor and response variables and is separated into training and test data for cross-validation. Regression methods apply algorithms to find associations between predictor and response variables in numerical training data and thereof produce black-box models to quantitatively predict a response variable. The effectiveness of a model is evaluated by a performance parameter that is computed from the error between the measured and predicted response variable (Bishop, 2006; James et al., 2013). A typical application in wastewater treatment is the forecast of an effluent or influent concentration with a regression model (supervised ML). However, most studies built predictive models based on similar or the same input parameters as ASM models. Thus, the potential to build more robust data-driven models by including further available parameters was not exhausted so far (Alejo, 2021).

In this study we transfer this idea to the outlined challenge of oxygen transfer modelling and use methods of supervised ML to predict α-factors based on long-term off-gas measurements and operating data of three WWTPs. Ideally, a model to predict the α-factor is based on parameters that describe the three phases involved in oxygen transfer. But many such parameters are not easily measurable in activated sludge (e.g., bubble size distribution, turbulence, surfactant concentration). On the other hand, secondary variables may correlate with conditions that impact the oxygen transfer such as changing wastewater properties due to stormwater runoff from combined sewers (Larsen et al., 1998; Schwarz et al., 2021; Wilén et al., 2006). Although the α-factor standardizes activated sludge water temperature and electrical conductivity, typical decrease due to stormwater inflow could represent rainfall events in a dataset in more detail. As a result, including operating data that has previously been overlooked in models has the potential to improve prediction performance.

For this study ex-situ off-gas measurements were performed for over three years to collect enough data to use ML methods. Oxygen transfer dynamics of various WWTP process designs were examined, including a two-stage AS treatment plant and two CAS WWTPs executed as a plug-flow reactor (PFR) and a closed-loop reactor (CLR). The objectives of this study were to (1) compare the range of α-factors in various WWTP process designs and discuss influence of typical wastewater and activated sludge characteristics on oxygen transfer; (2) examine the spatial variation of α-factor within activated sludge tanks especially due to upstream anoxic denitrification zones in PFR and CLR processes; and (3) predict α_0-factors with a machine learning model (Random Forest) based on operating data of WWTPs.

P.3.3 Methodology

P.3.3.1 Determination of α-factors with ex-situ off-gas columns

Oxygen transfer parameters and the α-factor were determined with pilot-scale test reactors performing ex-situ off-gas tests as described in ASCE/EWRI 18-18 (2018). The oxygen uptake rate (OUR) is measured within the ex-situ columns at steady-state conditions of dissolved oxygen (DO) by a mass balance of off-gas analysis and DO concentration to determine oxygen transfer parameters in activated sludge. A flow diagram of the process is shown in Figure P.3.1 Two aeration tanks with duplicate machinery and instruments were used to examine two AS zones simultaneously.

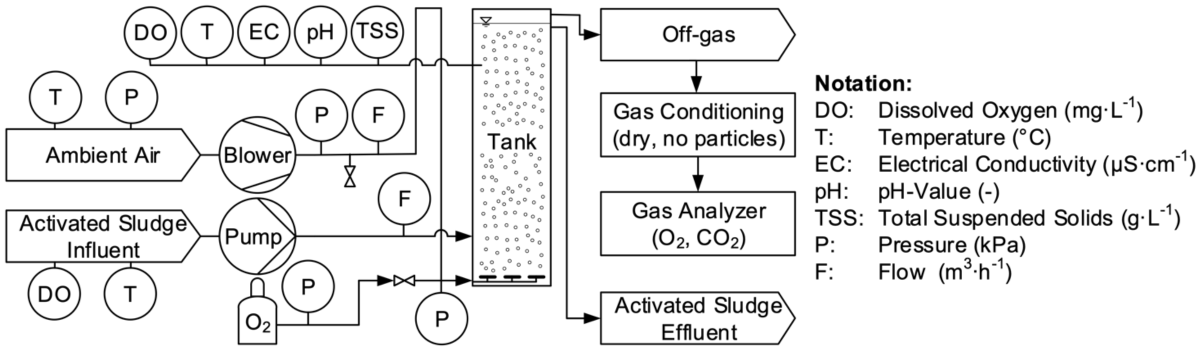

Figure P.3.1 Flow diagram of an ex-situ column for steady-state off-gas testing

A detailed description of the pilot reactors can be found in (Schwarz et al., 2022) and most notable characteristics are summarized below. Reactors of dimensions 1.2 m x 1.2 m x 5.8 m (L x W x H) with a volume of 8.3 m^3 were equipped with fine-bubble disc diffusers with a density of 13.5 %. Blowers were operated to set airflow rates (specified for tank volume) within a range of 0.5 to 2.5 $Nm^3 \cdot m^{-3} \cdot h^{-1}$. Sludge transfer was controlled to maintain a hydraulic retention time (HRT) of 15 minutes according to ASCE/EWRI 18-18. Pressure cleaning to remove biofilm growth on diffuser surface and reactor walls was performed monthly and reverse flexing of disc diffusers twice a week to reduce effects of scaling and fouling during long-term off-gas measurements. Therefore, the α-factor is reported without a fouling factor in this study. Twice a week, online sensors were cleaned, and gas analyzer was calibrated to ensure proper data recording of instruments. Clean water oxygen transfer measurements were performed three times at each examined WWTP and averaged to be used as a denominator to determine the α-factor. Pressure loss of diffusers during long-term measurements at each WWTP increased by about 1 kPa due to fouling and scaling. A comparison measurement has shown that the pilot-setup could record α-factors with a mean relative standard deviation of about ± 2.8 %.

P.3.3.2 Introduction of α_0-factor for off-gas testing under nonsteady-state conditions

The ex-situ method allows to operate the blowers of the ex-situ columns independently from the examined activated sludge tank. Usually, activated sludge is transferred from an aerated zone into an ex-situ column and the airflow rate is controlled to maintain DO at steady-state conditions to replicate conditions of in-situ off-gas measurements using off-gas hoods (ASCE, 2018; Boyle, 1983). DO in the ex-situ column is maintained at the same concentration as DO in the activated sludge tank zone (DO_{zone}) and the α-factor is determined as the ratio of standard oxygen transfer rate in process water ($SOTR_{pw}$ in $g \cdot h^{-1}$) and SOTR in clean water ($SOTR_{cw}$ in $g \cdot h^{-1}$):

$$\alpha = \frac{SOTR_{pw}}{SOTR_{cw}} \quad \text{(steady-state)} \qquad \text{(P3.1)}$$

Activated sludge can also be transferred from non-aerated tanks. This approach therefore allows to determine theoretical α-factors in anoxic or anaerobic tank zones. Under these nonsteady-state conditions DO in the activated sludge tank (DO_{zone}) and the ex-situ off-gas column differ. To compare α-factors in aerated and non-aerated zones the mass balance to determine the α-factor must be adjusted to consider DO_{zone} in the transferred sludge as follows:

$$\alpha_0 = \frac{SOTR_{pw} - (DO_{zone} \cdot Q_{AS})}{SOTR_{cw}} \quad \text{(nonsteady-state)} \qquad \text{(P3.2)}$$

This adjustment subtracts DO in the examined activated sludge tank zone (DO_{zone} in $g \cdot m^{-3}$) that is transferred to the ex-situ column (Q_{AS} in $m^3 \cdot h^{-1}$) from SOTR in process water ($SOTR_{pw}$ in $g \cdot h^{-1}$). When operating an ex-situ reactor with sludge from a non-aerated zone (zero DO_{zone}), no adjustment is made. This calculation produces α_0-factors that are generally lower than α-factors, especially the higher the oxygen concentration in the aeration tank is. Another difference is the oxygen diffusion gradient in the activated sludge tank and ex-situ reactor under nonsteady-state conditions. This difference is considered by standardizing the oxygen transfer efficiency to an oxygen transfer at a base of zero DO according to ASCE 18-18. Still, activated sludge from an aerated tank zone inevitably includes a gas phase while activated sludge from a non-aerated tank zone does not, which results in slightly higher α_0-factors in aerated zones than in non-aerated zones. To summarize, using the α_0-factor improves the comparison of oxygen transfer results between aerated and non-aerated zones considerably compared to the α-factor. α-factors based on off-gas measurements under steady-state conditions are still required to design aeration systems. This study exclusively presents results as the α_0-factor because of its focus on the oxygen transfer dynamics in different WWTP processes and activated sludge tank zones. For further details and a comparison of the α_0 and α-factor based on parallel measurements see supplementary information section P.3.7.1.

P.3.3.3 Overview of examined wastewater treatment plants and operating data

The pilot reactors were operated over a period of more than three years to perform long-term off-gas measurements on the sites of three different WWTPs. Process designs included a two-stage activated sludge system (WWTP A1 and A2) and two CAS systems designed as a plug-flow reactor (WWTP B) and a closed-loop reactor (WWTP C). Figure P.3.2 presents a flow diagram of the WWTPs with layout of biological treatment stages, clarifiers, and connecting wastewater and sludge flows such as return activated sludge (RAS), waste activated sludge (WAS), and internal recirculation. It also includes the position of the sludge transfer points across the tank length of each activated sludge tank as a percentage of its total length from inflow to effluent. In WWTP B and WWTP C α_0-factors were measured in different tank zones while in WWTP A1 and A2 only the inlet zone of each stage was measured.

The two-stage activated sludge plant is separated into a first high-rate activated sludge stage (A1, red) and a second stage for nitrification (A2, yellow). It has a bypass and recirculation flow and both stages have no internal recirculation. Nitrate is partially removed in an upstream denitrification zone of the second stage that is also fed with readily biodegradable substrate from the inflow via a bypass. The remaining nitrate is removed in a downstream denitrification (DN) filter. A full description of the two-stage WWTP A1/A2 can be found in (Schwarz et al., 2021). WWTP B (green) and C (blue) are both separated into anoxic, aerobic, and transition zones. The aeration system in the transition zone is controlled depending on ammonia effluent concentration and can be turned off completely. During dry weather WWTP B has a relative share of recirculation and RAS of about 82 % compared to the inflow and the closed-loop reactor of WWTP C is operated with agitators at an estimated circulation time of about 35 minutes to prevent sedimentation. About one third of wastewater inflow of WWTP B was from industrial sources, while WWTP A and C were primarily treating municipal wastewater from a combined sewer system.

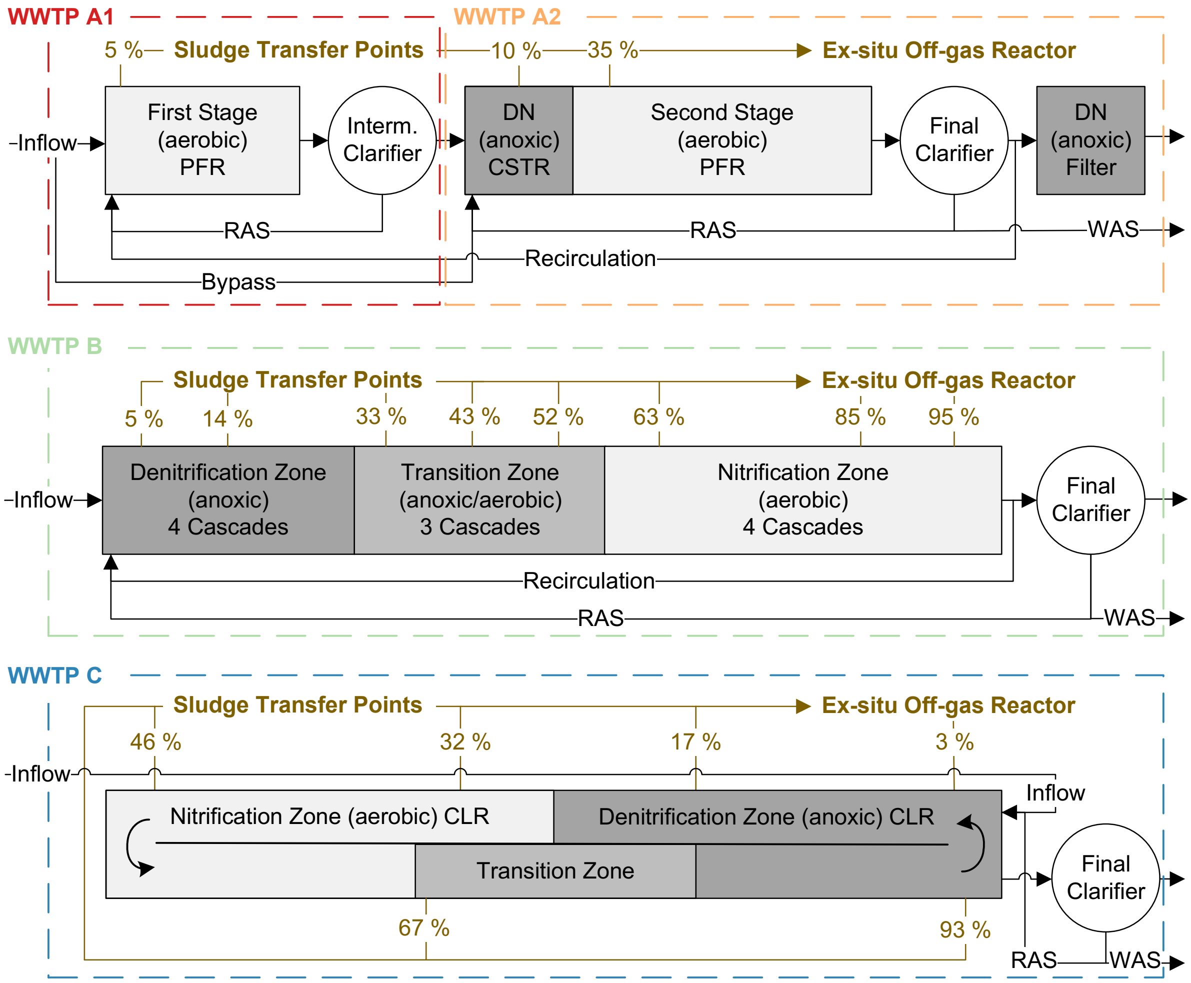

Figure P.3.2 Overview of examined WWTPs and sludge transfer points for ex-situ off-gas testing in each activated sludge tank

Table P.3.1 lists key operating data to further characterize the examined WWTPs in the upper part as well as operation of ex-situ columns and the amount of collected off-gas data in the lower part. Data is stated as approximate values (~) or as mean values ± standard deviation (SD).

The volume specific airflow rate in the activated sludge (AS) tank refers to the respective aerobic tank volume. Blowers in the ex-situ columns were operated to match the airflow rate in the AS tank for operation at WWTP A1 and A2. However, at the sites of WWTP B and C higher airflow rates were set in the ex-situ columns because operation in anoxic zones required higher oxygen transfer to exceed a DO concentration of 0.3 mg·L^{-1} required for off-gas testing. A limitation of the ex-situ off-gas method is that the airflow rate in the ex-situ column affects the determined α-factor (Schwarz et al., 2022). Therefore, a narrow range of volume specific airflow rates was set in the ex-situ tanks to avoid a bias of the α_0-factor. At each WWTP site off-gas data was collected over a period that included a seasonal shift of conditions and resultant changes of operation (e.g., water temperature, TSS, and SRT). Collected off-gas data was compressed to 1-hour intervals. This interval is suitable because under ideal mixing conditions at an HRT of 15 minutes in the ex-situ columns about 98 % of activated sludge is replaced within 1 hour (Schwarz et al., 2022).

Table P.3.1 Overview of key operating parameters of examined WWTPs and ex-situ column operation

Parameter	**Unit**	**WWTP A1**	**WWTP A2**	**WWTP B**	**WWTP C**
Population equivalent (PE)	-	> 1 Mio.	> 1 Mio.	~ 700,000	~ 250,000
Sludge retention time (SRT)	d	~ 2	~ 30	~ 15	~ 25
Wastewater inflow (dry weather)	$m^3 \cdot s^{-1}$	2.6	2.6	1.2	0.5
Maximum inflow (rain weather)	$m^3 \cdot s^{-1}$	7.0	7.0	4.0	1.2
Total org. carbon (TOC) inflow	$mg \cdot L^{-1}$	144 ± 37	46 ± 10	189 ± 59	141 ± 39
Total suspended solids (TSS)	$g \cdot L^{-1}$	2.2 ± 0.4	5.4 ± 0.5	4.0 ± 0.3	5.6 ± 0.5
Vol. spec. airflow rate (AS tank)	$Nm^3 \cdot m^{-3} \cdot h^{-1}$	1.8 ± 0.5	0.7 ± 0.2	0.7 ± 0.2	0.5 ± 0.1
Vol. spec. airflow rate (ex-situ)	$Nm^3 \cdot m^{-3} \cdot h^{-1}$	1.8 ± 0.2	0.9 ± 0.1	1.5 ± 0.1	1.0 ± 0.0
Period of measurement	months	13	13	11	7
Collected off-gas data	days	105	110	342	214

In total 17 parameters were chosen as predictor variables to train a machine learning model. All parameters were derived from continuous operating data from in-situ online sensors or ex-situ online analyzers. These typical instruments were used by WWTP operators for process control and routinely validated and calibrated via laboratory analysis. Data reconciliation was conducted according to the IWA Good Modelling Practice Guidelines (Rieger, 2012). In general, the predictor variables collected at all three WWTP sites represented the following criteria:

- wastewater influent concentrations (carbon as TOC, nitrogen as NH_4-N and NO_3-N, phosphorus as PO_4-P),
- mass balance of activated sludge tank with influent and effluent wastewater and sludge recirculation flows,
- state of the activated sludge process (airflow rate, DO, pH),
- standardization parameters (water temperature, electrical conductivity (EC), atmospheric pressure),
- position along the activated sludge tank.

Some of these basic parameters were further processed to include predictor variables that are typically used for oxygen transfer modelling or to consider measurement related effects. These feature engineering approaches included:

- calculation of TOC load and TOC F/M ratio because these parameters are common to describe oxygen transfer inhibition in activated sludge,
- dilution of wastewater inflow due to internal recirculation and return activate sludge (e.g., TOC represents the concentration in wastewater inflow whereas TOC_{in} considers all inflows of a treatment stage, i.e., supernatant of return activated sludge, internal recirculation, and bypass flows, proportional to their respective water flow),
- time shift to consider delay between online sensor measurement in inflow and off-gas monitoring in ex-situ column (especially when examining rear tank zones, see supplementary information section P.3.7.2).

To improve comparability the predictor variables were standardized where possible (e.g., by using a volume specific airflow rate instead of a total airflow rate). Nonetheless, these predictor variables cannot be identical between the WWTP datasets because operators used different sensors or calibration methods. For a full list of predictor variables with units, values ± SD and a short description see supplementary information section P.3.7.3.

P.3.3.4 Training of a Random Forest model to predict the α_0-factor

17 predictor variables as described above and listed in supplementary information section P.3.7.3 were used to train a machine learning model to predict the α_0-factor as a response variable. All predictor variables were derived from operating data as provided by the WWTP operators. The α_0-factor was measured with the ex-situ off-gas pilot reactors. The applied machine learning method was the Random Forest (RF) algorithm which is a decision tree-based method (Breiman, 2001). Random Forest trains an ensemble of decision trees which are created by randomly sampling from the training dataset (bootstrap sample). In addition, a split at each node can only use one predictor variable of a randomly selected subset. Repeating this procedure (recursive binary splitting) grows trees that are not pruned until a split creates a terminal node which cannot be split without falling below a minimum node size (number of remaining observations from the training dataset). Once the ensemble of trees is trained, the final prediction is made by aggregating an average of the predictions of all trees. Random Forest was chosen as a machine learning method for the α_0 prediction model because of its proven prediction performance in related fields and its capability to cope with interactions and correlations of predictor variables which are typical in operating data describing wastewater composition (Tyralis et al., 2019).

The dataset used in our study includes about 20,000 complete observations of predictor and response variables as 1-h intervals (divided for the three WWTPs as given in Table 1). 20 % of the data were randomly sampled from the dataset and retained as a test dataset to quantify the prediction performance of the developed Random Forest models. No additional data preprocessing was performed, as no transformation of distribution or normalization is required for the application of the Random Forest algorithm (Kuhn and Johnson, 2019). The number of trees in the ensemble (*ntree*) was set to 500 and the terminal node size (*nodesize*) to 5. The number of randomly selected predictor variables at each node (*mtry*) was determined by grid search tuning based on lowest RMSE (root-mean-square error) with a 10-fold cross-validation and 5 repeats. To determine the performance of the model the mean values of the parameters RMSE, R^2 (coefficient of determination) and MAE (mean absolute error) were determined.

Random Forest offers the possibility to determine the relative importance of predictor variables (Svetnik et al., 2003). However, these variable importance measures are biased towards correlated predictor variables (Strobl et al., 2008) which complicates design of a parsimonious Random Forest prediction model (Genuer et al., 2010). WWTP operating data contains many correlated variables, e.g., parameters characterizing wastewater contaminants. See supplementary information section P.3.7.5 for a correlation matrix of all predictor and response variables. Therefore, the number of predictor variables in the model is not reduced because of a potential bias in predictor variable selection. This has the advantage that model performance for each WWTP can be directly compared based on the same predictor variable set.

Instead of examining variable importance, we examined the effect of missing predictor variables on model performance in case a WWTP operator cannot use a parameter implemented in our prediction models. Each type of instrument defines a set of predictor variables. For example, dissolved oxygen sensors are required to determine DO in the examined tank zone as well as the average across all aerobic zones. Both predictor variables would be missing to train a Random Forest model, if no DO sensors were available at a WWTP. The method to consider missing instruments deleted a set of predictor variables, retrained and tuned the Random Forest model as described above, and then compared the model error of the reduced and original model. The model error was calculated as a percentage increase of RMSE of the reduced model with missing predictor variables and the original model with all predictor variables of Random Forest predictions on test data.

Statistics and visualization were done using R 3.6.3 (R Core Team, 2020), tidyverse package (v1.3.0) for visualization (Wickham et al., 2019), data.table package (v1.14.0) for data handling (Dowle and Srinivasan, 2021), caret package (v6.0-90) for feature selection, cross-validation and model training (Kuhn, 2021), and randomForest package (v4.7-1) to train regression based Random Forest models (Liaw and Wiener, 2002).

P.3.4 Results and Discussion

P.3.4.1 Overview of WWTP operation and resulting α_0-factors

The activated sludge stages of the examined WWTPs differed in process layout, operation, and wastewater composition which resulted in a different range of α_0-factors (see Figure P.3.3B). A principal component analysis (PCA) was performed on a subset of 8 predictor variables to visualize their correlation in one diagram (Figure P.3.3A). The first two principal components (PC1 and PC2) cover a total variance of 65 % and predictor variables are represented as arrows (loadings). Loadings pointing in the same direction are positively correlated and vice vera. The data points represent the operating data where an α_0-factor was recorded with the ex-situ off-gas method. 5,000 randomly sampled data points of each WWTP are colored to distinguish the WWTPs and complemented by three density lines for each WWTP containing 25 %, 50 % and 75 % of the available data. Overall, PCA shows four clusters of operating data which visualizes that many predictor variables do not overlap between WWTP datasets. As an example, the high-rate activated sludge WWTP A1 was operated at significantly higher airflow rates and TOC F/M ratios, whereas the second stage was operated at higher DO setpoints. Figure P.3.3B shows the resulting α_0-factors as an empirical cumulative distribution diagram of the whole dataset. The high-rate activated sludge system A1 and its second stage A2 had the lowest and highest range of α_0-factors, respectively. Overall, WWTPs with clusters close to each other in the PCA also had α_0-factors in a similar range.

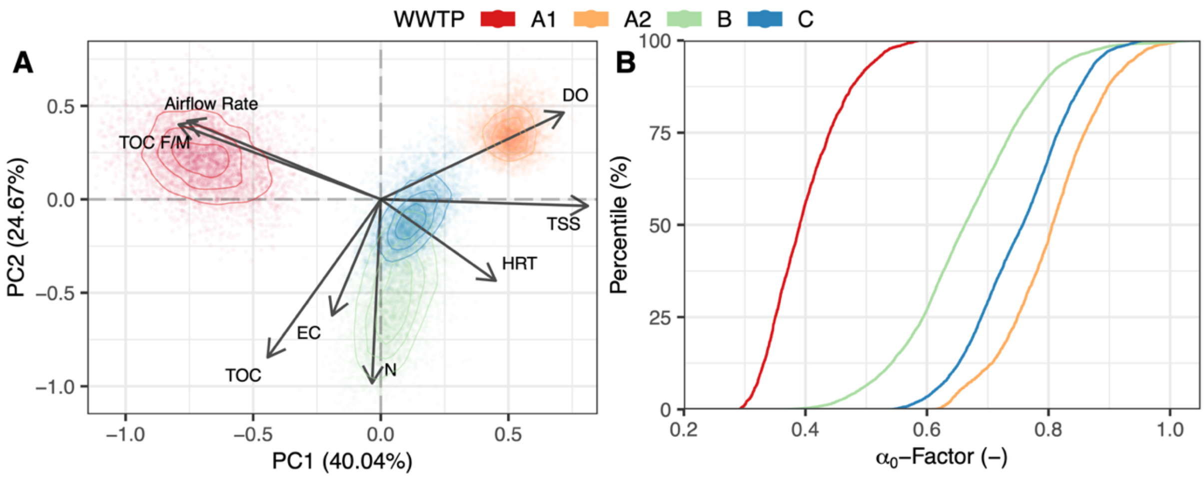

Figure P.3.3 PCA of operating data showing clusters of WWTPs (3A) and empirical cumulative distribution of resulting α0-factors (3B)

Although our data represents the examined WWTP's operation in practice, Figure P.3.3 shows that the available datasets are fragmented and therefore insufficient to develop an oxygen transfer prediction model that is generally applicable to all types of WWTPs. Instead, individual models for each WWTP were trained to analyze if a prediction of the α_0-factor based on operating data was possible.

P.3.4.2 Spatial and diurnal variation of α_0-factor

Ex-situ off-gas measurements were performed in aerated and non-aerated tank zones to determine α_0-factors in successive tank zones of three WWTPs divided into anoxic, transition, and aerobic zones at different intervals of tank length. The data presented in Figure P.3.4 was collected only during dry weather conditions for better comparison and the amount of data is indicated by a number below the boxplots. On average, α_0-factors increased across the tank length in all reactor types. In WWTP B a significant increase was only detected after the initial inflow zone whereas subsequent zones showed similar ranges of α0-factors. Removal of wastewater contaminants by biosorption and biodegradation in an activated sludge tank across the tank length reduced oxygen transfer inhibition causing a spatial variation of the α0-factor. For the first time, this effect could also be confirmed by off-gas measurements in anoxic zones, which suggests an anoxic removal that improved oxygen transfer in down-stream aerated zones. Based on the data the respective impact of biosorption and biodegradation on oxygen transfer improvement cannot be differentiated. The concept of spatial variation of α-factors, although known for decades, has just recently been included by Bencsik et al. (2022) in dynamic oxygen transfer modelling. The composition of the wastewater-sludge matrix within an activated sludge tank is rarely monitored, however, the position in a tank is a possible surrogate parameter to describe spatial variation. Therefore, the tank zone is also included as a predictor variable in our machine learning approach.

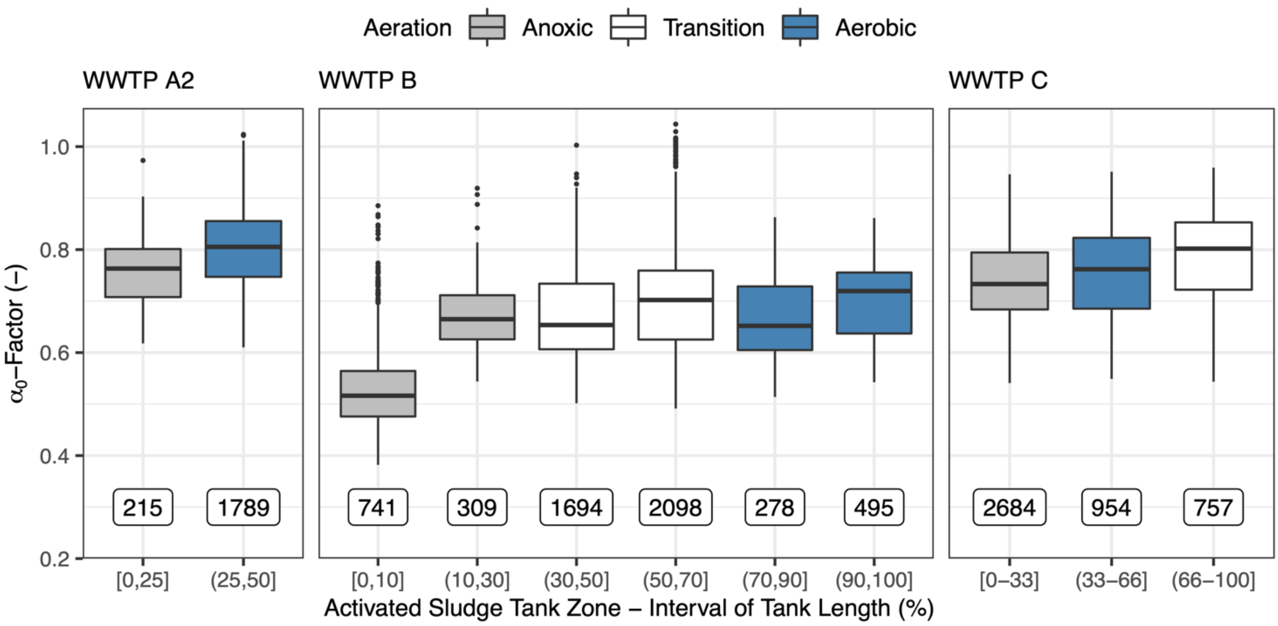

Figure P.3.4 Spatial variation of α_0-factor along the activated sludge reactors of WWTP A2, B and C with number of 1-h observations below each boxplot

In addition to spatial variation, α_0-factors of the examined WWTPs followed a diurnal cycle, see Figure P.3.5. For WWTP A1 (red) and A2 (yellow) α_0-factors in the aerobic inflow zone of the aeration tank are plotted. For WWTP B (green) and WWTP C (blue), in addition to the anoxic inflow zone, all subsequent aeration and transition zones are summarized in one diagram (other zones). The lines represent the course of the percentiles from 5th to 95th percentile, the dashed line represents the mean value throughout the day. The lines were determined as smoothed functions of the percentiles or mean values of 15-min interval data. The peak α_0-factors were measured during the daytime rather than at night due to the long residence time of wastewater in the sewer network.

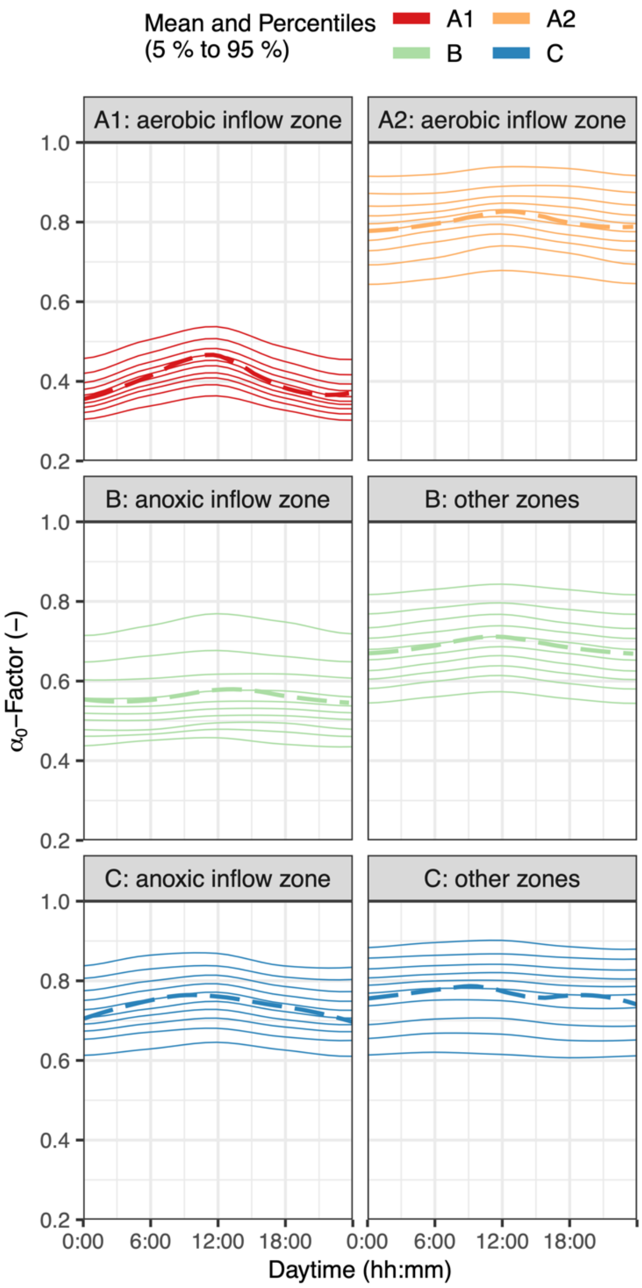

Figure P.3.5 Diurnal variation of α_0-factor

WWTP A1 shows a distinct peak of the α_0-factor, whereas diurnal variation of α_0-factor in the other WWTPs had a more uniform pattern with a less pronounced peak. In

WWTP A2 the influent load was reduced and buffered by the upstream high-rate activated sludge stage (A1) and the upstream denitrification zone. In WWTP B and particularly in the closed loop reactor of WWTP C influent wastewater was diluted by internal recirculation flows which led to a similar pattern in the anoxic inflow zone and the subsequent zones. Overall, this confirms that the higher the internal recirculation is within an activated sludge tank, the flatter the diurnal variation is due to a distribution of the wastewater load (Rosso and Stenstrom, 2007).

In our machine learning approach time of day was not included as a predictor variable to describe the diurnal pattern of α_0-factor. Bencsik et al. (2022) have shown that a sinusoidal pattern of α-factors fails to predict oxygen transfer correctly during stormwater events. Instead, in our machine learning approach predictor variables such as TOC_{in} were used to consider dilution effects that differ during dry and wet weather conditions.

P.3.4.3 Influence of wastewater load on α_0-factor

Chemical oxygen demand (COD) is a key parameter to calibrate oxygen transfer prediction models as wastewater load changes dynamically during a day (Bencsik et al., 2022; Jiang et al., 2017). In our study, we used TOC to describe influent wastewater characteristics, because ex-situ TOC online analyzers are common in larger WWTPs and produce a higher temporal resolution than COD laboratory analysis. Similarly, an inverse relationship between TOC F/M ratio and α-factor exists (Günkel-Lange, 2013).

Figure P.3.6 compares α_0-factor with TOC F/M ratio and its individual components, i.e., the actual hydraulic retention time in activated sludge tanks including recirculation flows (HRT_a), TOC influent concentration including recirculation flows (TOC_{in}) and total suspended solids (TSS). The TOC F/M ratio is plotted on a logarithmic scale. Data points are colored and surrounded by density lines as in Figure P.3.3 to distinguish the examined WWTPs.

The inverse relationship between α-factor and wastewater influent load has been described in previous studies (Jiang et al., 2017; Leu et al., 2009). For WWTP operators, this negative correlation means that oxygen transfer is generally lowest when oxygen demand is highest during high load. Our collected results from four different activated sludge stages confirm this for TOC_{in} (Figure P.3.6A) and TOC F/M ratio (Figure P.3.6D). However, within one WWTP this relationship is superimposed by various other impacts resulting in a less clear relationship. It is worth emphasizing that Figure P.3.6 presents the available datasets used for machine learning based model development but is not suitable to derive simple linear regression models from it.

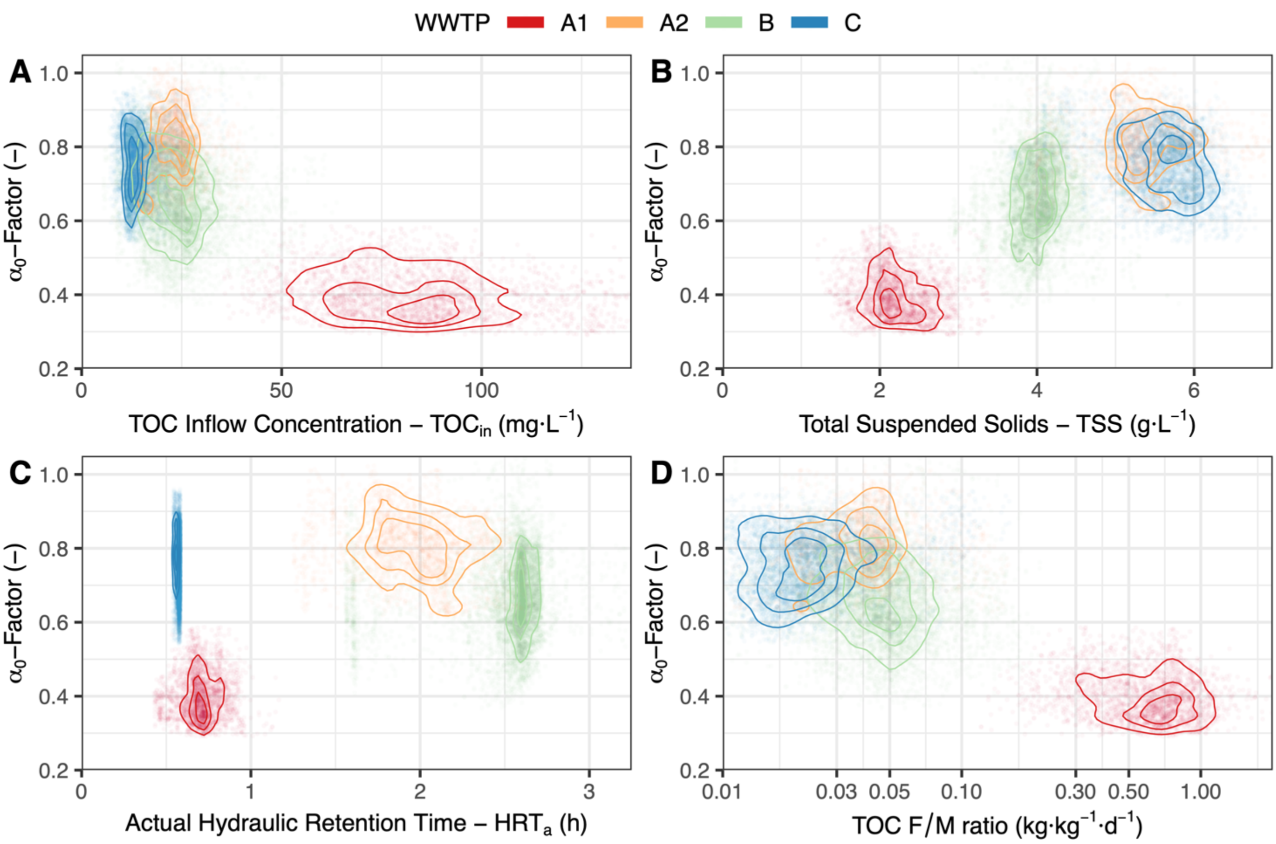

Figure P.3.6 Comparison of α_0-factor with predictor variables TOC_{in} (6A), TSS (6B), HRT_a (6C), and the thereof aggregated parameter TOC F/M ratio (6D)

A positive relationship between α-factor and TSS was proposed by Baquero-Rodríguez et al. (2018). They argued that higher TSS improves oxygen transfer due to increased biosorption of oxygen transfer inhibiting substances on sludge flocs. Although our results show this positive relationship between α_0-factor and TSS concentrations up to 6 $g \cdot L^{-1}$ (Figure P.3.6B), this influence could not be examined isolated from other impacts. Overall, the relevance of this relationship in one individual activated sludge tank remains unclear, as TSS is usually kept within a narrow range during operation.

The high-rate activated sludge system A1 is operated at significantly lower HRT_a than the other WWTPs (Figure P.3.6C). Increased wastewater inflow during rain lowers HRT_a, but a distinct impact on α_0-factor cannot be spotted in our data. HRT_a is not a suitable parameter to describe all wastewater characteristics that change oxygen transfer during stormwater inflow. Models should distinguish first flush with highly loaded wastewater from diluted stormwater.

Overall, the bivariate diagrams in Figure P.3.6 demonstrate that no individual parameter can be used to reliably predict the α_0-factor. Instead, a multivariate approach should be used to model oxygen transfer to consider various superimposed impacts and interaction effects. Below, we present such a data-driven methodology.

3.4 Predicting the α_0-factor with Random Forest

Four separate Random Forest models were trained to predict α_0-factors based on 17 predictor variables. Figure P.3.7 shows a comparison of the α_0-factors predicted with the Random Forest models and the α_0-factors measured with the ex-situ off-gas method from the test dataset. Diagrams on the left are separated into the four activated sludge stages and scaled within the same range. WWTPs are further distinguished by color and a dashed black linear regression is added in each diagram. A density distribution of the prediction error is shown on the right.

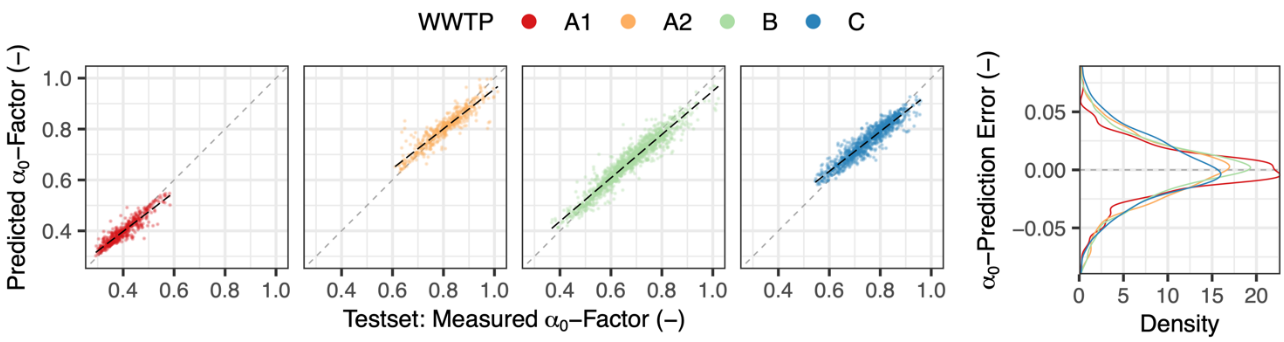

Figure P.3.7 Comparison of predicted α_0-factors with test dataset

Random Forest successfully predicted the α_0-factor for all WWTP processes. The regression line indicates that in each case the prediction of high α_0-factors was underestimated, and low α_0-factors were overestimated. This regression towards the mean value occurs because prediction results are averaged at each terminal node in training and across the ensemble of trees for each prediction by Random Forest algorithm. In addition, in the upper and lower ranges of the α_0-factor fewer training data was available for model training, thus potentially reducing model performance. A learning curve confirms that model accuracy would benefit from even more training data (see supplementary information section P.3.7.6).

An important constraint of many data-driven methods is that they are unable to generalize, meaning that extrapolation for conditions that are not covered by the training dataset is difficult to accomplish. Consequently, the presented Random Forest models would lose their predictive power once the WWTP operation or wastewater composition is altered significantly from the observed training data. Nonetheless, the high potential of oxygen transfer predictions with a machine learning method is demonstrated by our results, as these are based on long-term measurements that include rain weather conditions and seasonal variation. Furthermore, α_0-prediction error is in the same range as the density distribution in Figure P.3.7 shows, even though different WWTP process layouts and resultant α_0-factors were examined. The Random Forest performance parameters are listed in Table P.3.2. Better model accuracy is indicated by lower RMSE and MAE and higher R^2.

Table P.3.2 Overview of Random Forest performance parameters RMSE (root-mean-square error), MAE (mean absolute error), and R2 (coefficient of determination)

WWTP	RMSE	MAE	R^2
A1	0.024	0.017	0.855
A2	0.033	0.024	0.840
B	0.033	0.023	0.911
C	0.031	0.022	0.880

In our previous study we discussed the measurement uncertainty of ex-situ off-gas testing and quantified the standard deviation of measurement uncertainty for a series of comparison measurements in WWTP B to ± 0.018 (Schwarz et al., 2022). A prediction performance undercutting the measurement error would suggest an overfitting model. In our case, the determined RMSE of 0.031 of the Random Forest model for WWTP B is in the range of the error of the ex-situ off-gas method suggesting that prediction performance could be limited by the measurement uncertainty of the off-gas method.

Predictor variables were selected because they are typically available to monitor the activated sludge process at WWTPs. Some predictor variables correlate with the α_0-factor (see supplementary information section P.3.7.5). Even without such a correlation, a parameter could be relevant to consider interaction effects. The Random Forest algorithm does not require careful selection of predictor variables and can cope with correlated predictor variables (Guyon and Elisseeff, 2003). It allows to include parameters that have not been used to predict oxygen transfer before such as water temperature and atmospheric pressure. Hence, the model potentially considers relevant information that has previously been ignored when modelling oxygen transfer.

In practice, a type of instrument used in this study might not be available to include as a predictor variable for model development. In some cases, this can reduce the model performance as Figure P.3.8 shows. It illustrates the percentage increase of RMSE if a Random Forest model is trained without a specified set of predictor variables.

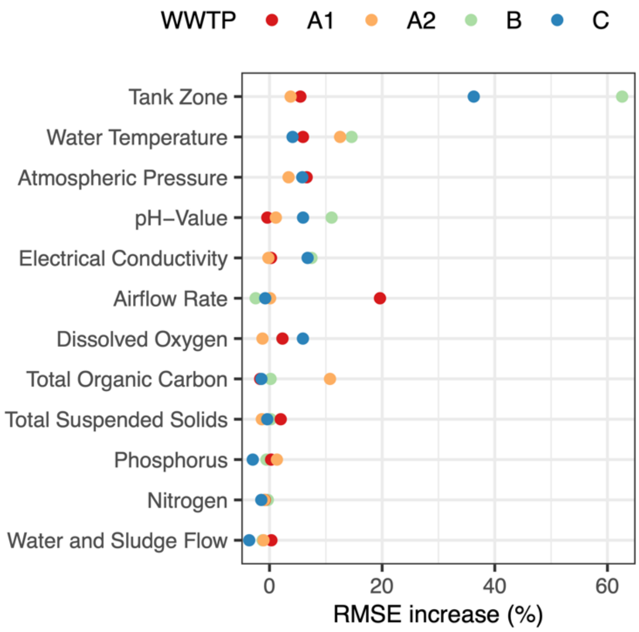

Figure P.3.8 RMSE increase of RF models with different sets of missing predictor variables compared to a RF model with complete set of predictor variables

For example, not including the predictor variable *tank zone* in a Random Forest model for WWTP B increases RMSE by more than 60 % compared to the Random Forest model with all predictor variables, thus reducing overall model performance. The importance of some predictor variables differs between WWTPs. Most notably, the spatial variation of α_0-factor as described by *tank zone* is only relevant in models of WWTP B and C that were trained with data from various tank zones (see Figure P.3.4). Likewise, *tank zone* was not relevant in WWTP A1 and A2 because only a small section of the tanks was covered with off-gas measurements (see Figure P.3.2). In some instances, predictor variable sets that are highly correlated with each other (e.g., TOC, N, P) improved model performance if left out. Here, a more parsimonious model improved RMSE by up to 4 %. A similar improvement was possible with recursive feature elimination (see supplementary information section P.3.7.4).

Overall, this comparison reveals that some of the standardization parameters (e.g., water temperature, atmospheric pressure, electrical conductivity) provided relevant information to improve a Random Forest based α_0-prediction model. In contrast, parameters usually used in oxygen transfer modelling (e.g., DO, TOC, water and sludge flow) were less important due to high correlation. Including all of them did not benefit model accuracy, therefore, a prediction model could still be developed even though certain instruments were not available at a WWTP. It is worth noting that Figure P.3.8 only demonstrates which parameters were important for the model predictions in the trained Random Forest models and cannot evaluate the actual relationship between parameters and α_0-factor. Overall, the application of the machine learning based prediction of oxygen transfer is not limited due to missing instruments and sensors to record predictor variables but because off-gas testing to monitor the activated sludge tank is rarely implemented in WWTPs.

P.3.4.4 Comparison with the current state of dynamic oxygen transfer modelling

The dynamic prediction of α-factors is a recent focus in the field of aeration technology as dynamic α prediction models by Bencsik et al. (2022) or Jiang et al. (2017) show. Instead of relying on static values, dynamic models can improve design and operation of aeration systems as site-specific influences on oxygen transfer are considered. However, it was not possible to develop a generally applicable oxygen transfer model yet. Previously reported models required calibration and validation based on site-specific full-scale WWTP data to predict oxygen transfer, similarly to our approach. Consequently, the machine learning model presented in this study could not fill this gap either but has unique advantages during model development. Some examples include:

- Previously reported dynamic oxygen transfer models relied on sludge retention time (SRT) to develop or calibrate model parameters. SRT is often difficult to determine reliably and especially at high temporal resolution varies significantly when waste activated sludge is withdrawn intermittently (Balbierz and Knap, 2017). In addition, Rosso et al. (2005) demonstrated the broad range of average α-factors for a given SRT at numerous WWTPs. Instead of relying on the definition of a WWTP's SRT, our method can consider all parameters involved in SRT as individual predictor variables of a model.
- A kinetic model based on municipal wastewater would have to be extended by new model components in case of industrial wastewaters containing significant amounts of non-biodegradable surfactants (Bencsik et al., 2022). In contrast, a ML model is automatically adjusted to site-specific wastewater characteristics if these are included in a training dataset.
- A ML model could consider effects of fouling and scaling of diffusers on oxygen transfer by including diffuser pressure loss as a predictor variable in a time series analysis. This effect was not examined in our study as regular maintenance of diffuser membranes prevented excessive fouling during the experiments.
- Selection and engineering of predictor variables could involve additional site-specific parameters (e.g., dosage of precipitation chemicals) that could further improve prediction performance.

P.3.5 Conclusions

A novel oxygen transfer model was presented by applying machine learning methods to predict the α_0-factor. The data-driven approach was based on long-term ex-situ off-gas testing that was conducted on three WWTPs, including a two-stage and two CAS systems with different reactor types. The key findings are outlined below:

- The spatial and diurnal variation of the α_0-factor was confirmed in various reactor types, which included ex-situ off-gas measurements in non-aerated anoxic zones for the first time. We therefore introduced the α_0-factor to compare off-gas data from aerated and non-aerated activated sludge tank zones measured with pilot-scale ex-situ off-gas reactors. This suggests that biosorption and biodegradation of wastewater contaminants in upstream denitrification zones increase α_0-factor in successive aerated zones.
- Random Forest models to predict the α_0-factor were trained exclusively with online operating data available to the WWTP operators and did not require extensive laboratory analysis. This multivariate approach considered wastewater characteristics, treatment plant operation, standardization parameters, and the spatial variation within an activated sludge tank as predictor variables that were not included in oxygen transfer modelling before.
- Model prediction was reliable with an RMSE between 0.024 and 0.033 (R^2 between 0.84 and 0.92) even though the examined activated sludge stages differed regarding their wastewater characteristics or operation. The Random Forest models dynamically predicted α_0-factors for regular WWTP operation, during stormwater events, and seasonal variation. Like previously reported dynamic oxygen transfer models, the machine learning methodology could not create models to reliably predict oxygen transfer under conditions not included in training data.
- The machine learning approach we presented in our study did not require calibration of model parameters. The methodology simplifies model development if enough data is available to train a data-driven model and therefore benefits from big datasets. We reported results for α_0-factors to include theoretical spatial variation within the whole activated sludge tank. The methodology is also applicable to predict α-factors in aerobic stages.

Overall, this article invites operators of wastewater treatment facilities to implement continuous off-gas monitoring in the activated sludge process. Even though it is possible to develop an initial model based on temporarily conducted off-gas testing, highest accuracy can be accomplished when continuous off-gas monitoring is implemented in the activated sludge tank. Multi-objective optimizations with a predictive instead of reactive control of the aeration system would benefit from such a model. A prospective application of machine learning based oxygen transfer prediction models could be in WWTPs with digital twins and supervisory control strategies with multiple control loops for activated sludge tanks. Collecting off-gas data today is going to enable operators to implement better aeration control strategies tomorrow.

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P.3.7 Supplementary Information

P.3.7.1 Comparison of α_0 and α-factor

The ex-situ off-gas method enables to examine oxygen transfer in aerated and non-aerated activated sludge tank zones. The α_0-factor considers the different oxygen transfer when sludge with or without a gas phase is transferred to the ex-situ reactors by an adjusted mass balance. Figure P.3.9 shows a direct comparison of the two types of α-factors. In this parallel measurement, one ex-situ reactor was operated in a non-aerated zone (α_0 or α-factor on abscissa) and one ex-situ reactor in an adjacent aerated zone (α_0 or α-factor on ordinate). The determined α-factors are shown in yellow and the α_0-factors in blue and are divided into three diagrams for different WWTPs. Linear regression lines are added for each of the two calculations. Note the different axis scales of the diagrams because of the different ranges of α-factors of the three WWTPs.

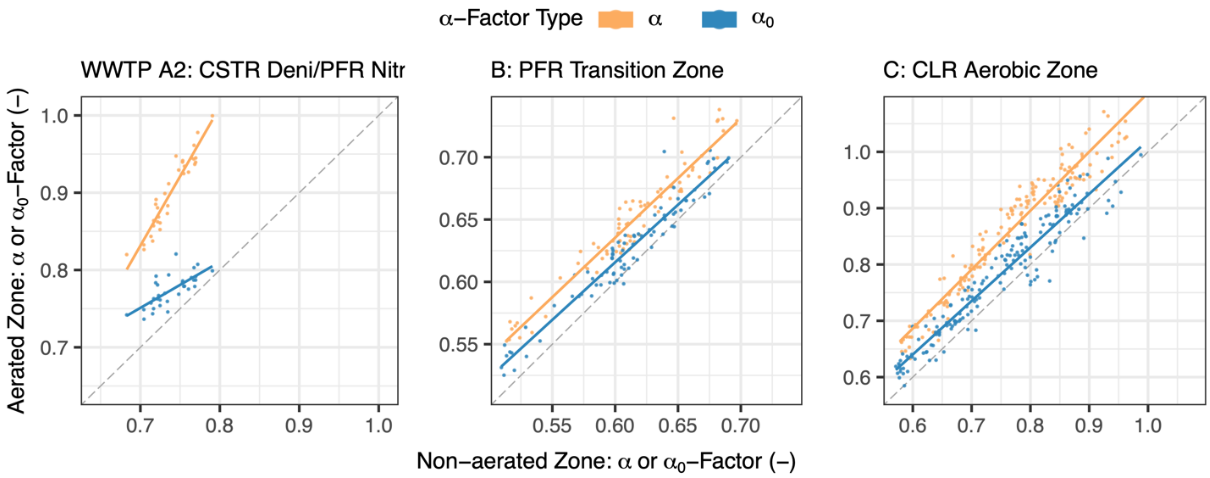

Figure P.3.9 Comparison of α_0 and α-factor in aerated and non-aerated zones

If the oxygen transfer in the adjacent tank zones does not change significantly, a distribution of the measured α-factors along the angle bisector (gray dashed line) is to be expected. For both the α_0-factor and the α-factor, the measured values are higher in the aerated zone than in the non-aerated zone. However, this effect is more pronounced for the α-factor. The adjustment of the mass balance at the α_0-factor thus improves comparability of the oxygen transfer in aerated and non-aerated zones compared to the α-factor without adjustment. Nevertheless, a systematic difference between α_0-factors from aerated and non-aerated zones remains. For WWTP A2 (upstream CSTR denitrification zone followed by PFR nitrification zone) and WWTP B (two consecutive cascades in the transition zone), a non-aerated zone was compared with a downstream aerated zone. In WWTP C (aerobic zone of a closed loop reactor), a non-aerated zone followed an aerated zone in the flow direction of the loop reactor. In both cases, higher α_0-factors are measured in aerated zones than in non-aerated zones. Accordingly, a possible change in the wastewater activated sludge matrix (e.g., due to further degradation of substances that inhibit oxygen transfer) between the adjacent aeration tank zones is not primarily causing different α_0-factors. The remaining difference between α_0-factors in aerated and non-aerated zones results from the intake of the gas phase into sludge transfer hoses, which distinguishes aerated from non-aerated zones. Nonetheless, Figure P.3.9 shows that using the α_0-factor improves the comparison of oxygen transfer results between aerated and non-aerated zones considerably.

P.3.7.2 Time shift: delay between sensor measurement in inflow and ex-situ off-gas monitoring

Many online sensors and ex-situ analyzers are operated in the influent or effluent of an activated sludge (AS) tank. In contrast, ex-situ off-gas measurements can be conducted across the whole AS tank length by positioning sludge transfer hoses in various tank zones. In plug-flow reactors (PFR) a delay results between a measured influent or effluent parameter and the α-factor from off-gas measurements. This temporal delay should be considered when predicting the oxygen transfer because wastewater composition is subject to a diurnal change.

Figure P.3.10 shows an AS tank as PFR schematically as an arrow from influent to effluent. Somewhere in between the influent and effluent parameters an α-factor can be measured with an ex-situ off-gas column. Depending on the hydraulic retention time (HRT) in the AS tank and the position of off-gas measurement the time shift to be considered changes. Figure P.3.10 exemplarily shows that an α-factor measured at the middle of an AS tank that has an overall HRT of 120 minutes at dry weather requires a time shift of t_{-60} for influent parameters and t_{60} for effluent parameters. In this case an α-factor measured at time t_0 is correlated with parameters that were measured in the influent 60 minutes earlier (t_{-60}) and effluent parameters that were measured in the effluent 60 minutes later (t_{60}).

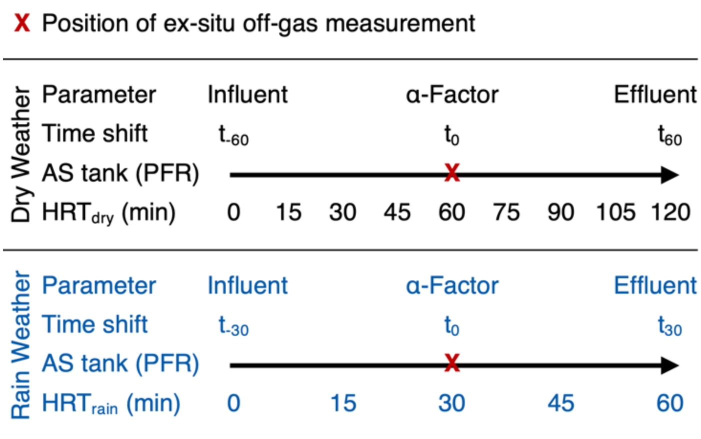

Figure P.3.10 Time shift between influent/effluent parameters and α-factor at dry and rain weather

Consequently, HRT in the AS tank changes when wastewater inflow changes. This is especially relevant when HRT is reduced by stormwater inflow. As shown exemplarily in the lower part of Figure P.3.10 (in blue), HRT is reduced by half and thus the time shift to be considered changes with it. To account for this time shift in the training datasets we shifted operating data according to HRT in intervals of 15 minutes. Figure P.3.11 shows an α-factor and parameters measured at the position of sludge transfer (ASP) in a table. Data obtained at t_0 are marked by an "n", while earlier data at t_{-15} is marked with "n-1", etc. In this example influent (IP) and effluent parameters (EP) were recorded 30 minutes earlier or later (HRT of ± 30 min). Thus, the α-factor and ASPs at t_0 are linked with IPs at t_{-30} and EPs at t_{30} and so on.

Time	α	ASP_1	...	ASP_i	IP_1	...	IP_i	EP_1	...	EP_i
t_{-45}					n-1	n-1	n-1			
t_{-30}					n	n	n			
t_{-15}	n-1	n-1	n-1	n-1	n+1	n+1	n+1			
t_0	n	n	n	n						
t_{15}	n+1	n+1	n+1	n+1				n-1	n-1	n-1
t_{30}								n	n	n
t_{45}								n+1	n+1	n+1

Figure P.3.11 Tabular example of time shift linking measured data of α-factor with activated sludge parameters (ASP), influent parameters (IP) and effluent parameters (EP) for a HRT of ± 30 min

P.3.7.3 Overview of predictor variables to train a Random Forest model to predict the α_0-factor

Table P.3.3 gives an overview of the predictor variables used to train a Random Forest model to predict the α_0-factor. The α_0-factor as the response variable of Random Forest regression is listed below the predictor variables. The table lists mean values alongside the sample standard deviation (SD) during standard operation of the WWTP. Additionally, the 5th and 95th percentiles are given to describe minimum and maximum conditions of WWTP operation that are only exceeded infrequently.

Table P.3.3 Overview of predictor variables and response for model training

Predictor/ Response Variables	Set	Description	Unit	WWTP A1 (First Stage)		WWTP A2 (Second Stage)		WWTP B		WWTP C	
				mean ± SD	5th – 95th percentile	mean ± SD	5th – 95th percentile	mean ± SD	5th – 95th percentile	mean ± SD	5th – 95th percentile
DO Zone	1/12	Dissolved oxygen in the examined AS tank zone	$mg{\cdot}L^{-1}$	0.3 ± 0.3	0.1 - 0.9	2.5 ± 1.2	0 - 3.4	0.6 ± 0.5	0 - 1.5	0.5 ± 0.6	0.1 - 1.9
DO	1	Mean dissolved oxygen in aerobic zones of AS tank	$mg{\cdot}L^{-1}$	0.6 ± 0.3	0.2 - 0.9	3.2 ± 0.2	3.1 - 3.4	1.3 ± 0.3	1 - 1.7	1 ± 0.6	0.3 - 2.0
AFR	2	Volume specific airflow rate in aerobic AS Tank zones	$Nm^3{\cdot}m^{-3}{\cdot}h^{-1}$	1.8 ± 0.5	0.9 - 2.3	0.7 ± 0.2	0.5 - 1	0.7 ± 0.2	0.3 - 1.1	0.5 ± 0.1	0.4 - 0.7
Q_{in}	3	Volume specific wastewater inflow	$L{\cdot}m^{-3}{\cdot}h^{-1}$	416 ± 183	191 - 832	189 ± 72	93 - 376	46 ± 34	18 - 142	40 ± 18	22 - 86
HRT_a	3	Actual hydraulic retention time in AS tank (excl. clarifier)	h	0.7 ± 0.1	0.5 - 0.9	2 ± 0.3	1.4 - 2.4	2.5 ± 0.3	1.7 - 2.7	0.6 ± 0.0	0.5 - 0.6
N	4	NH_4-N and/or NO_3-N influent concentration considering time shift	$mg{\cdot}L^{-1}$	36.2 ± 10.1	16.7 - 51.6	28.5 ± 7.7	14.8 - 41.6	65.9 ± 22.9	26.7 - 98.8	46.5 ± 11.9	19.9 - 63.4
P	5	PO_4-P influent concentration considering time shift	$mg{\cdot}L^{-1}$	3.6 ± 0.9	1.8 - 4.9	2.1 ± 0.9	0.8 - 3.7	9.5 ± 3.5	3.6 - 15.2	4.3 ± 1.5	1.5 - 6.4
TOC	6	TOC influent concentration considering time shift	$mg{\cdot}L^{-1}$	144 ± 37	89 - 217	46.3 ± 9.9	26.3 - 60.5	189 ± 59	83 - 281	141 ± 39	74 - 210
TOC_{in}	3/6	TOC_{in} concentration in influent considering time shift and dilution	$mg{\cdot}L^{-1}$	83.7 ± 20.9	52 - 125	22.9 ± 4.9	15.4 - 30.9	23.9 ± 8.3	13.2 - 38.6	13.9 ± 3.4	9.9 - 20.9
TOC load	3/6	TOC load in influent considering time shift	$t{\cdot}d^{-1}$	32.9 ± 14.6	12.7 – 58.7	13.6 ± 5.8	6.3 – 25.6	15.9 ± 8.7	6.5 – 31.9	2.5 ± 1.4	1.1 – 5.4
TOC FM	3/6/7	TOC F/M ratio in influent considering time shift	$g{\cdot}kg^{-1}{\cdot}d^{-1}$	641 ± 274	252 - 1089	41 ± 22	17 - 89	49 ± 27	20 - 98	25 ± 18	10 – 61
TSS	7	Total suspended solids (TSS)	$g{\cdot}L^{-1}$	2.2 ± 0.4	1.6 - 2.9	5.4 ± 0.5	4.2 - 6.3	4 ± 0.3	3.6 - 4.6	5.6 ± 0.5	4.6 - 6.3
p_{atm}	8	Atmospheric pressure	mbar	1010 ± 11	992 - 1028	1013 ± 10	994 - 1030	1013 ± 9.5	996 - 1028	1015 ± 11	995 - 1029
T_w	9	Water temperature in activated sludge	°C	18.3 ± 2.5	13.3 - 21.2	18.5 ± 3	13.6 - 22.1	18.2 ± 3	13.4 - 23	14.5 ± 1.4	12.2 - 16.9
EC	10	Electrical conductivity	$\mu S{\cdot}cm^{-1}$	1255 ± 226	749 - 1539	1106 ± 190	698 - 1309	1404 ± 347	714 - 1849	1218 ± 217	718 - 1465
pH	11	pH in activated sludge	–	7.6 ± 0.2	7.4 - 8	7.3 ± 0.1	7.1 - 7.4	7 ± 0.2	6.6 - 7.3	6.9 ± 0.2	6.6 - 7.1
α_0-factor	–	Recorded with ex-situ off-gas columns	–	0.40 ± 0.06	0.31 - 0.52	0.80 ± 0.08	0.65 - 0.94	0.66 ± 0.11	0.48 - 0.84	0.75 ± 0.08	0.61 - 0.88

An additional predictor variable (*Tank Zone, Set 12*) is the position of sludge transfer as a percentage of tank length in flow direction (compare with Figure P.3.2 in main text). At this position the dissolved oxygen DO_zone is measured. The predictor variable *Tank Zone* considers that oxygen transfer dynamics change locally depending on operation of AS tank zones (anoxic/aerobic) and continuous biodegradation of wastewater contaminants that inhibit oxygen transfer. In total, 17 predictor variables were selected for ML model training. In the trained Random Forest models, no effluent parameters were used as predictor variables because information about future effluent conditions cannot be implemented in a predictive model for real-time applications.

The column "Set" marks all predictor variables that were recorded with the same type of sensor or instrument. For example, both predictor variables *DO* and *DO zone* require dissolved oxygen sensors in the activated sludge tank and are therefore in the same set. These sets were defined to determine importance of certain instruments to develop Random Forest models for prediction of the α_0-factor.

0.6 % of the final dataset was missing that included all relevant predictor variables and the α0-factor as response variable. k-nearest neighbors (KNN) as a multiple imputation technique was used to impute the missing values (k = 5, Euclidean distance, scaled data) (Kuhn and Johnson, 2013; Troyanskaya et al., 2001).

TOC concentration in influent was measured by ex-situ online analyzers. The parameter had a low signal to noise ratio (SNR). It was therefore additionally smoothed by applying a 3-h moving average. This parameter was then used in all other TOC related predictor variables. All other predictor variables remained as 1-h intervals of the operating data.

P.3.7.4 Algorithm for recursive feature elimination with external resampling

Random Forest offers the possibility to determine the relative importance of predictor variables (features) (Svetnik et al., 2003). Based on feature importance a subset of features can be determined to design a parsimonious Random Forest prediction model (Genuer et al., 2010; Guyon and Elisseeff, 2003). The algorithm below was used for recursive feature elimination (RFE) with the training dataset to build a parsimonious Random Forest model. RFE implemented an external resampling step for predictor variable selection to improve model performance on new observations that considered the variability caused by predictor variable selection. The selection of predictor variables was determined based on the best model performance as measured by the lowest RMSE of the RF models with a 10-fold cross-validation and 5 repeats. Training of the Random Forest models was then performed with the selected subset of predictor variables.

```
For each resampling iteration do
    Partition data into training and validation set via resampling
    Train the RF model on the training set with all features
    Predict the validation set data
    Calculate feature importance
    For each feature subset Fi with i = 1…n do
        Keep Fi most important variables
        Train the model on the training set with Fi predictors
        Predict the validation set data
    End
End
Calculate the RMSE performance for all Fi for the validation data
Determine the appropriate number of features by lowest RMSE
Fit the final model based on the optimal number of features Fi for the original training set
```

Modified from "The caret Package" (Kuhn, 2019), https://topepo.github.io/caret/index.html (last access 2022/05/20); see section 20 "Recursive Feature Elimination" for further details on code implementation.

Table P.3.4 compares performance parameters of Random Forest models with all 17 predictor variables and the models after recursive feature elimination. In these 9 to 12 predictor variables remained (Rem. Var.) and RMSE improved by up to 7 % compared with the base models.

Table P.3.4 Comparison of Random Forest performance parameters with all predictor variables and remaining variables after RFE

WWTP	RMSE	R^2	MAE	RMSE	R^2	MAE	Rem. Var.
	All Predictor Variables			Recursive Feature Elimination			
A1	0.024	0.856	0.017	0.023	0.866	0.017	12
A2	0.033	0.840	0.023	0.032	0.849	0.022	9
B	0.031	0.922	0.022	0.029	0.929	0.020	9
C	0.030	0.886	0.023	0.028	0.891	0.022	9

P.3.7.5 Correlation matrix of response and predictor variables

Diagrams below show correlation matrices of response and predictor variables using the non-parametric Kendall's rank correlation. The non-parametric approach was used instead of parametric Pearson's rank correlation because some parameters were not normally distributed and not all relationships between variables were linear.

Variables as described in Table P.3.3 are listed at the horizontal and diagonal axis. For each pair of variables, the correlation is indicated by a colored shape. Shapes stretch from a high-contrast line marking a high correlation near ± 1 to a pale circle marking a low correlation near 0. Colors range from red for negative correlations (- 1 to 0) to blue for positive correlations (0 to +1) as indicated by the vertical axis.

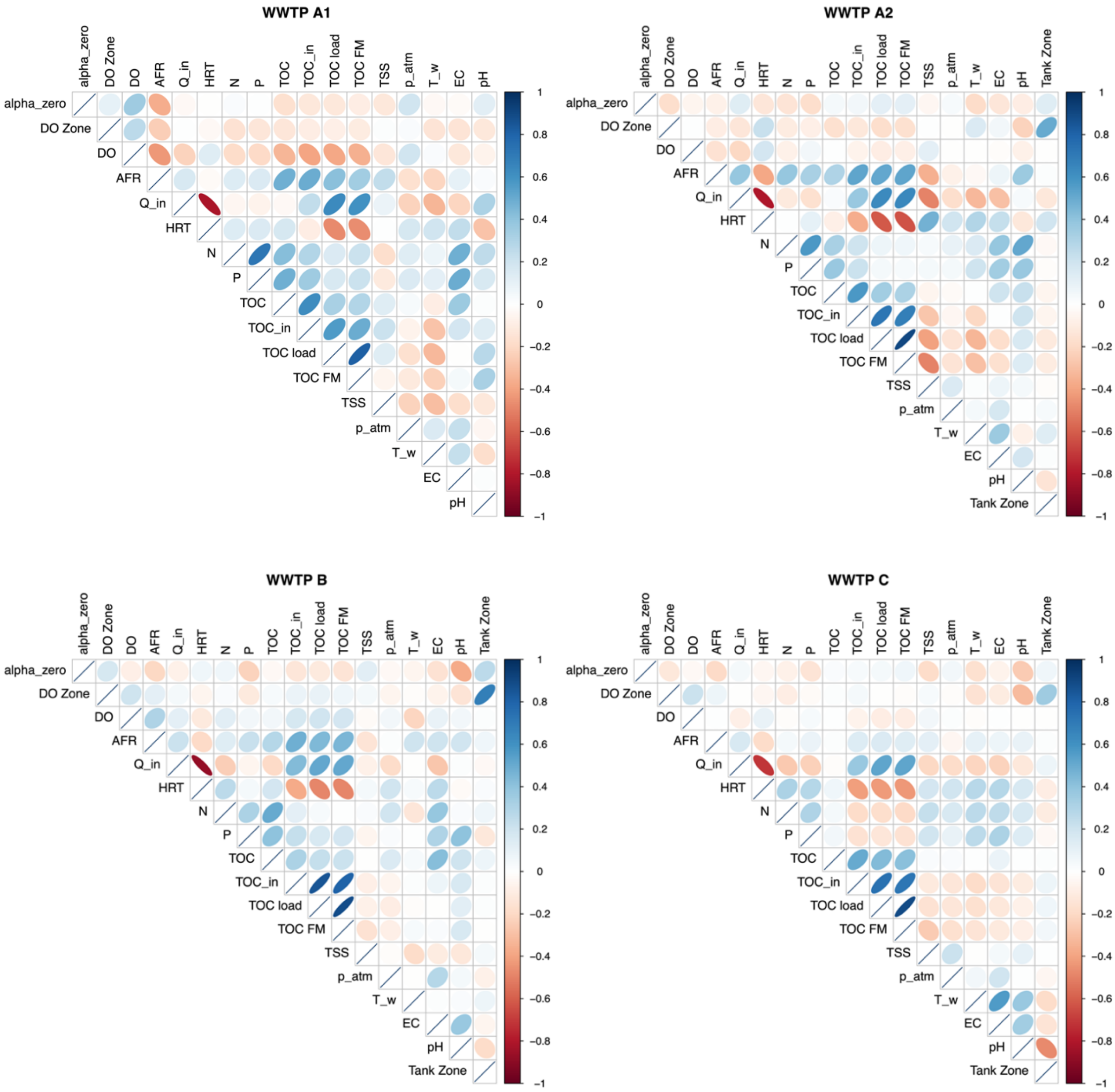

Figure P.3.12 Correlation matrices of response and predictor variables for all examined WWTPs

P.3.7.6 Learning Curve

Figure P.3.13 shows learning curves of Random Forest models for each WWTP distinguished by color. The three diagrams are divided into RMSE, MAE, and R^2 to visualize the model's prediction performance if only smaller datasets were available for model training. The performance parameters at 100 % of the training dataset equal the values reported in Table P.3.2 of the main text for the models using all available off-gas data for model training. Random Forest models were trained with random subsamples of the available training data to examine how the prediction performance changes when fewer off-gas measurements were available as training data. Random Forest models were trained with these restricted datasets but with the same hyperparameters as the full model as reported in section P.3.3.4 of the main text. As the share of the training dataset decreases, lower prediction performances were determined. For example, R^2 was significantly lower when using only 25 % of the available data to train a Random Forest model. Or else, the more training data was available for model training the better the prediction performance was. The learning curves are shaped as saturation curves that have not converged to the saturation value. This suggests that the presented methodology would benefit from even larger datasets to develop an even more accurate prediction model for the α_0-factor.

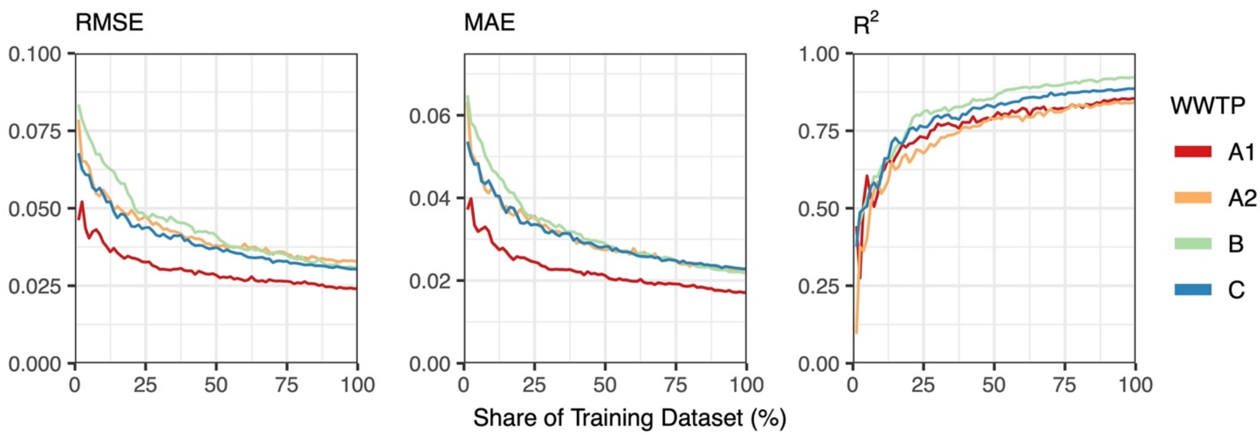

Figure P.3.13 Learning curves for RF models of each WWTP divided into RMSE, MAE, and R^2

P.3.7.7 Training and test performance parameters of Random Forest models

Table P.3.5 compares the performance results for training and test datasets. The model results for test datasets are reported in the main document in Table P.3.2 as these represent the performance of models expected for new input data collected under the same conditions as the training dataset. The performance results for training datasets are generally better than for test datasets due to overfitting of the Random Forest models to the training dataset. The higher the difference between training and test performance parameter the more overfitting of the model to operating conditions only present in the training dataset occurred. The spread of results in Table P.3.5 suggest that some overfitting is still present. Potential solutions to reduce this overfitting include further model tuning or selection of a different supervised machine learning method for model training.

Table P.3.5 Overview of Random Forest performance parameters

WWTP	RMSE Training	RMSE Test	MAE Training	MAE Test	R^2 Training	R^2 Test
A1	0.010	0.024	0.007	0.017	0.979	0.855
A2	0.013	0.033	0.010	0.024	0.978	0.840
B	0.014	0.033	0.010	0.023	0.986	0.911
C	0.012	0.031	0.009	0.022	0.983	0.880

P.3.7.8 Additional references in supplementary information

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5 Conclusions and Outlook

The focus of my dissertation was to examine oxygen transfer dynamics in activated sludge and the potential of oxygen transfer modelling based on a data-driven machine learning approach. Summarized below are the key conclusions and prospective applications of each paper individually. Papers in this cumulative work follow a logical sequence. Starting with an in-depth analysis of the ex situ off-gas method used throughout this work (P1) and followed by a study of oxygen transfer in two-stage systems as a special variant of activated sludge processes (P2), the dissertation concludes with the introduction of a data-driven prediction of oxygen transfer in various activated sludge systems (P3). However, papers in this cumulative work were not published in this sequence. P2 was published before P1 because a two-stage WWTP was examined at first. P1 was later published when a sufficient data basis was available that included more typical oxygen transfer conditions in a CAS system. Therefore, sections below also show how the papers are linked with each other and describe findings that were recognized in hindsight. Finally, an outlook summarizes next steps to improve our understanding of oxygen transfer dynamics and its modelling.

Paper 1

Novel insights how to properly conduct and evaluate ex situ off-gas measurements were published in P1. Oxygen concentration in the off-gas was determined as the most important input quantity for reliable off-gas measurement exceeding all other parameters. Off-gas collection and analysis should therefore be given special attention when designing pilot setups. An uncertainty analysis revealed that measurement uncertainty of ex situ off-gas tests to determine the α-factor can be lower than previously reported measurement uncertainty of ± 5 to 10 %. This theoretical approach can be used to evaluate the measurement uncertainty of pilot setups a priori. P1 also demonstrated that α-factors recorded with the ex situ method are systematically increased by additional mixing in the pilot reactor through the lateral sludge inflow. Consequently, oxygen transfer results obtained with the ex situ method differ from results of the in situ method and should clearly be stated as ex situ derived results.

Overall, my findings in P1 provide complementary information missing in technical guidelines regarding the ex situ off-gas method that is crucial for future applications of the ex situ off-gas method. In general, findings regarding the estimation of measurement uncertainty are transferable to the in situ off-gas method because the same type of instruments are used in both methods.

Paper 1 and Paper 2

Static α-factors to design aeration systems in two-stage processes were quantified in P2. However, P1 demonstrated that results from ex situ and in situ measurements cannot be compared because lateral sludge inflow systematically increases α-factors in the ex situ setup. This limitation of the ex situ method was unknown when P2 was published because the in-depth analysis of the methodology was conducted later as part of P1.

As a conclusion, the α-factors reported in P2 were too high although the experiments were explicitly conducted as described in ASCE/EWRI 18-18 (2018). Future experiments to determine α-factors for design purposes should therefore use in situ off-gas measurements.

Paper 1 and Paper 3

I discussed several conditions and constraints of the ex situ off-gas method in P1 that I had to consider to develop oxygen transfer models and validate their results in P3.

Firstly, a direct comparison of measurements under the same conditions exemplarily demonstrated that α-factors recorded with the pilot setup have a relative standard deviation of about ± 2.8 %. This provided an important threshold that allowed me to further validate results in P3. Here, a prediction of α-factors undercutting the measurement uncertainty of the ex situ method would indicate overfitting of the underlying machine learning model. This was not the case with the Random Forest method presented in P3. It is advisable to estimate the measurement uncertainty of the response variable in the underlying training data to validate performance parameters of machine learning models.

Secondly, I discussed that repeating off-gas measurements is recommended due to the methodology's inherent measurement uncertainty and systematic measurement errors that can be caused, e.g., by fouling of diffusers, biofilm growth, sensor drift, or imperfect clean water testing. It is possible that a Random Forest model detects systematic measurement errors in a specific measurement period, thus overfitting to the training data. Therefore, the oxygen transfer modelling approach using Random Forest models as presented in P3 should use training data from long-term off-gas measurements spanning multiple months for each examined WWTP.

Thirdly, P1 showed the systematic bias of α-factors due to lateral sludge inflow depending on airflow rate in the ex situ reactor. Consequently, α-factors recorded at different WWTPs and airflow rates could not be compared in a universal model. Therefore, individual models were trained for each activated sludge stage of the examined WWTPs in P3.

Paper 2

In P2, α-factors for design load cases were determined as 0.45 for α_{mean} and 0.33/0.54 for $\alpha_{min}/\alpha_{max}$ in the first stage (HRAS), and as 0.80 for α_{mean} and 0.69/0.91 for $\alpha_{min}/\alpha_{max}$ in the second stage. The absolute values of these α-factors would differ in situ as described above. However, P2 demonstrated for the first time that oxygen transfer dynamics differ tremendously in the two stages of a two-stage activated sludge process. In addition, a decrease of α-factors was observed in the first stage during wet weather conditions, thus suggesting that oxygen transfer is more susceptible to changing inflow characteristics in HRAS than in CAS systems. My observations in P2 generally confirmed the inverse relationship between TOC and the α-factor. Analysis of inflow surfactant concentrations showed that surfactant load was significantly lower in the second stage than in the first stage. Lower α-factors in the first stage could be attributed to the oxygen transfer inhibiting effect of surfactants but not quantified specifically for surfactants compared with TOC in general. Overall, considering these findings can improve the design of aeration systems in two-stage processes.

Paper 2 and Paper 3

Long-term off-gas measurements in P2 covered a typical range of WWTP operation conditions including seasonal variation, as well as dry and wet weather conditions. A range of α-factors was measured in both stages as expressed by the derived α-factors for design load cases. Even though general relationships between the α-factor and TOC were confirmed, the total variation of measured values could not be explained by simple correlations with operating parameters. It is important to recognize that many interacting factors influence oxygen transfer in the activated sludge at the same time. Based on this analysis, I concluded that a modelling approach based on multiple predictor variables would be required to advance modelling the oxygen transfer which led to my work presented in P3.

Paper 3

A machine learning approach was used to model oxygen transfer dynamics for the first time in P3. The data-driven method was developed and tested based on long-term ex situ off-gas testing conducted on a variety of WWTP process layouts, including a two-stage and two CAS systems with different reactor types. Additionally, the ex situ method allowed to observe potential influences on oxygen transfer throughout the whole activated sludge tank including non-aerated zones. I introduced the α_0-factor to examine spatial variation of oxygen transfer inhibition in aerated and non-aerated activated sludge tank zones and, thus, include the characteristics of these process layouts in my analysis.

Most oxygen transfer models designed so far are limited to few parameters, usually focusing on the influence of SRT, COD, and TSS on oxygen transfer (see section 2.3). The Random Forest models were trained with 17 predictor variables which included parameters such as water temperature, atmospheric pressure, and electrical conductivity that were previously only used for standardization of oxygen transfer parameters. Therefore, this approach allows to include additional parameters which can describe the conditions affecting oxygen transfer in the activated sludge tank that have been overlooked so far. Most importantly, the method only relies on operating data as predictor variables for model training that WWTP operators already measure.

The machine learning model predictions for the different activated sludge stages ranged between an RMSE of 0.024 and 0.033 (R^2 between 0.84 and 0.92). Whether this prediction performance is considered good enough depends on the prospective application and cannot be evaluated conclusively within this work. With my findings in P3 I want to encourage the aeration research community to include more parameters than “the usual suspects” to develop oxygen transfer models in the future.

Outlook

In recent years, the use of whole plant models has increased to enable process engineers to examine interactions between unit processes (Rieger, 2012). Oxygen transfer in the activated sludge is not only affected by the inflow wastewater characteristics, but also by operation of the activated sludge process and other influent flows from sludge treatment processes. Consequently, operators are confronted with multiple options to adjust the treatment process, each potentially affecting oxygen transfer in the activated sludge tank. In such a multi-objective optimization problem the implementation of a comprehensive oxygen transfer prediction model is required to find solutions and control the aeration system of an activated sludge tank accordingly. In this case, continuous off-gas measurements could improve monitoring of aeration systems and a dynamic prediction of α-factors based on a machine learning model could be utilized to control aeration systems as part of a digital twin in the future. To accomplish this, further research should focus on the aspects detailed below.

- *Model input data:* Information about influences on oxygen transfer contained in operating data is limited as described in section 2.2. For example, point measurements by online sensors can inevitably only represent local conditions within an activated sludge tank depending on the mixing conditions. Information about oxygen transfer inhibiting wastewater contaminants is only collected in influent and effluent flows and not available at the actual point of impact, the bubble rising in the activated sludge. In addition, some parameters oversimplify potential influences on the oxygen transfer. For example, the sum parameter COD cannot distinguish different types of wastewater contaminants such as surfactants that can differently inhibit oxygen transfer or TSS concentration estimated by a turbidity sensor cannot distinguish organic from inorganic matter or particle size distribution. Continuous development of sensor capabilities, increasing the number of measurements, and improving sensor reliability could add further information relevant to describe oxygen transfer dynamics in the future.
- *Model output data:* Oxygen transfer efficiency under process conditions is based on off-gas tests. So far, off-gas analyses have been used in numerous studies, but

application of gas analyzers is not part of the typical instrumentation on WWTPs. The implementation of in situ off-gas measurements in full-scale is required to model oxygen transfer for a WWTP, because no generally applicable model exists so far. Continuous off-gas testing would also allow to adjust models once wastewater or process characteristics of a treatment plant change. Moreover, more accurate off-gas measurements would reduce uncertainty about the state of oxygen transfer and enable more precise model development. Overall, better model input and output data describing and quantifying the oxygen transfer would benefit the development of mechanistic and machine learning models.

- *Model performance and reliability:* The supervised machine learning approach presented in this work is often unable to generalize under conditions not included in training data. Therefore, prediction errors could be caused by defective sensors or changing process characteristics. Hence, future applications in practice should consider strategies to increase resilience to a change of the system that could result in a faulty prediction of oxygen transfer. With my work I want to invite researchers to replicate the machine learning methodology with existing off-gas datasets from long-term measurement campaigns. Next steps include the comparison and coupling of mechanistic models with machine learning models and the implementation of an oxygen transfer prediction model into an aeration control scheme.

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