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Biodegradation half-lives of biodiesel fuels in aquatic and terrestrial systems: A review

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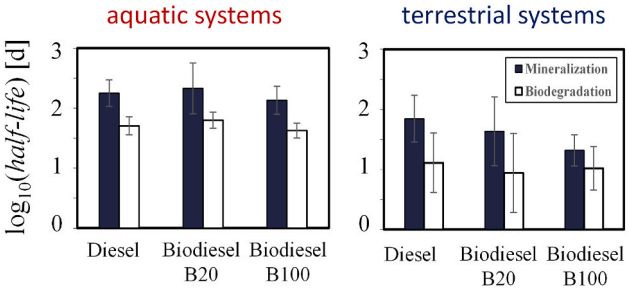
HIGHLIGHTS

- Systematic review of biodegradation kinetics of biodiesel fuels.
- Across all data points, biodegradation half-lives ranged from 9 to 62 days.
- Mineralization half-lives were 2–5.5 times longer than biodegradation half-lives.
- The half-lives were 2.5–8.5 times longer in terrestrial than in aquatic systems.
- Improved risk and impact assessment of biodiesel fuels.

GRAPHICAL ABSTRACT



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ABSTRACT

Information on biodegradation kinetics of biodiesel fuels is a key aspect in risk and impact assessment practice and in selection of appropriate remediation strategies. Unfortunately, this information is scattered, while factors influencing variability in biodegradation rates are still not fully understood. Therefore, we systematically reviewed 32 scientific literature sources providing 142 biodegradation and 56 mineralization half-lives of diesel and biodiesel fuels in various experimental systems. The analysis focused on the variability in half-lives across fuels and experimental conditions, reporting sets of averaged half-life values and their statistical uncertainty. Across all data points, biodegradation half-lives ranged from 9 to 62 days, and were 2–5.5 times shorter than mineralization half-lives. Across all fuels, biodegradation and mineralization half-lives were 2.5–8.5 times longer in terrestrial systems when compared to aquatic systems. The half-lives were generally shorter for blends with increasing biodiesel content, although differences in number of data points from various experiments masked differences in half-lives between different fuels. This in most cases resulted in lack of statistically significant effects of the type of blends and experimental system on biodegradation half-lives. Our data can be used for improved characterization of risks and impacts of biodiesel fuels in aerobic aquatic and terrestrial environments, while more experiments are required to quantify biodegradation kinetics in anaerobic conditions. Relatively high biodegradability of biodiesel may suggest that passive approaches to degrade and dissipate contaminants in situ, like monitored natural attenuation, may be appropriate remediation strategies for biodiesel fuels.

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1. Introduction

Biodiesel, a type of fuel derived mainly from plants, is blended with petroleum diesel oil in proportions ranging from 2 to 20%, depending on the country or region (Aktas et al., 2010; Bücker et al., 2011; Cadillo-Benalcazar et al., 2021; DeMello et al., 2007; Gupta, 2020; Schleicher et al., 2009). In the European Union, the Renewable Energy Directive (RED) for the period of 2010–2020 aimed at replacing 10% of fossil fuels with biofuels (Bórawski et al., 2019; Stattman et al., 2018; The European Parliament, 2009). Biofuel production has been steadily rising, with over the 10-fold increase in annual global biodiesel production between 2000 and 2020, reaching 45 billion litres in 2020 (IEA, 2020). This increase was met with growing awareness about potential negative consequences of production of biofuels from crop plants, mainly caused by the conversion of natural land to cultivated land which releases carbon bound in the natural biomass and the soil as CO_2, and competition with food and feed which (next to social impacts) may indirectly contribute to a loss of natural land somewhere else (Bjørn et al., 2018; Majer et al., 2009; Searchinger et al., 2008). These concerns resulted in regulations limiting biofuel production form 1st generation feedstock (*i.e.*, from crop plants) up to 7% in 2015 and ultimately 3.8% in 2030 (Stattman et al., 2018; The European Parliament, 2018), as well as restricting utilization of carbon-rich environments (*e.g.*, forests, wetlands, peat bogs) for crop farming (Bórawski et al., 2019; Stattman et al., 2018). At the same time, production of 2nd generation fuels (*e.g.*, from miscanthus, straw and reeds), as more environmentally sustainable when compared to biodiesel from 1st generation feedstock, was promoted (Bórawski et al., 2019).

Biodiesel is generally considered as readily biodegradable in the environment, with significantly higher biodegradation rates reported for blends with higher biodiesel content (Chen et al., 2019; Horel and Schiewer, 2011; Lisiecki et al., 2014; Makareviciene and Janulis, 2003; Owsianiak et al., 2009; Woźniak-Karczewska et al., 2019; Yassine et al., 2013). The term 'biodegradation' is defined as a 'process by which organic substances are decomposed by microorganisms (mainly aerobic bacteria) into simpler substances' (*OECD,* 1992). Mineralization (also referred to as ultimate biodegradation) can be defined as 'the level of degradation achieved when the test compound is totally utilised by microorganisms resulting in the production of carbon dioxide, water, mineral salts and new microbial cellular constituents (biomass)' (*OECD,* 1992).

Various factors influence biodegradation and mineralization kinetics of biodiesel fuels, including type of environment (aqueous, terrestrial) and exposure conditions (aerobic, anaerobic), or types of degrading microorganisms (autochthonous or introduced) (Chen et al., 2019, 2020; Dos Anjos et al., 2018; Lapinskiene and Martinkus, 2007; Soares Junior et al., 2009; Wieczorek et al., 2015; Woźniak-Karczewska et al., 2019). Selection of appropriate remediation strategy toward biodiesel fuels and assessment of environmental risks and impacts of biodiesel emissions to environment requires knowledge about these factors as well as data on biodegradation and mineralization kinetics in various conditions and types of environment (Owsianiak et al., 2023). One review on biodiesel's environmental fate has been published (Gupta, 2020), but it focuses on mechanisms of biodiesel transport in subsurface rather than on aspects related to biodegradation and mineralization. Individual studies presenting experimental results on its biodegradation and/or mineralization are available, but this data has not been systematically collected, analysed and compared, until now (Chen et al., 2020, 2019; Corseuil et al., 2011; Cruz et al., 2020; Cyplik et al., 2011; DeMello et al., 2007; Demirbaş, 2009; Dos Anjos et al., 2018; Horel and Schiewer, 2014, 2011, 2020, 2016; Lapinskiene et al., 2006; Lapinskiene and Martinkus, 2007; Li et al., 2016a; Lisiecki et al., 2014; Makareviciene and Janulis, 2003; Meyer et al., 2014; Montagnolli et al., 2019; Ng et al., 2015; Owsianiak et al., 2009; Pasqualino et al., 2006; Pinto Mariano et al., 2008; Schiewer and Horel, 2017; Silva et al., 2012; Soares Junior et al., 2009; Thomé et al., 2014; Vauhkonen et al., 2011; Wieczorek et al., 2015; Woźniak-Karczewska et al., 2019; Yassine et al., 2013; Zhang et al., 1998).

In this review, we compile data on biodegradation kinetics and elucidate factors which influence biodegradation and mineralization half-lives. We consider various media (*e.g.*, soils of different characteristics, artificial porous matrices, artificial effluents, and river water), aeration conditions, and type of biodiesel blends. Comparisons are made with petroleum diesel fuel, taken as a reference.

2. Review procedure

The review was conducted in two steps. In the first step, we identified all studies that address any aspect of biodegradation of biodiesel fuels and thus may contain data on biodegradation and/or mineralization kinetics. Those studies were identified in Scopus database, using combinations of keywords: *biodiesel, diesel, blend, biodegradation, mineralization, bacteria, degradation, microbial degradation*. Thereby, we identified 145 peer-review articles. In the second step, each of these articles was then searched for data on biodegradation or mineralization kinetics, like half-lives or rate constants. Data were included in the analysis if the following criteria were met: (1) half-life ($\tau_{1/2}$) values were either provided by authors or could be calculated from other data presented in the article (typically, rate constants); (2) type of fuel was specified; (3) type of system (aquatic, terrestrial) was specified and (4) aeration conditions were specified. Data on mineralization or biodegradation kinetics of diesel fuels were included only if measured in the same experiment as for biodiesel fuels. Experiments quantifying biodegradation of specific fuel components were not included as they could not be compared with overall biodegradation data, *e.g.*, chromatograms or mass fractions for biodiesel components, FAMEs composition evaluation. Approaches to express degradation as decrease in purity or as a change in indicator colour were also not included, as were data for blends which used other fuels than petroleum diesel fuel (*e.g.*, jet fuels, crude oil or pure petroleum hydrocarbons). With this, a total of 32 articles were found to contain data on mineralization and/or biodegradation kinetics which met the inclusion criteria of the second step of the review procedure.

To support interpretation of results, non-mandatory information with respect to other aspect that might affect biodegradation process, *e.g.*, specification of fuel, microorganism's origin (autochthonic microorganisms present in sample or prepared inoculum), soil moisture, and temperature was also reported, if available.

2.1. Biodegradation and mineralization half-lives

Most of the considered 32 studies reported biodegradation/mineralization extent of biodiesel fuels at given point time in time. Actual biodegradation/mineralization rate constants (k) or half-lives ($\tau_{1/2}$) were reported less frequently. In the majority of studies first-order kinetics applied:

$$C(\mathrm{t}) = C_0 \bullet \exp\,(-k \bullet t) \tag{1}$$

where $C(t)$ (various units, *e.g.*, mg/kg, mg/L) is the residual biodiesel fuel concentration, C_0 (various units, *e.g.*, mg/kg, mg/L) is the initial biodiesel fuel concentration and t (day) is the biodegradation or mineralization time. Rearranging and solving for k (day^{-1}) gives:

$$k = \frac{\ln[C_0] - \ln\,[C(t)]}{t} \tag{2}$$

from which we finally obtain half-lives $\tau_{1/2}$ (day):

$$\tau_{1/2} = \frac{\ln 2}{k} \tag{3}$$

If zero-order kinetics applied, the following equations were used:

$$C(t) = C_0 - k \bullet t \quad (4)$$

Rearranging and solving for k (concentration unit·day^{-1}) gives:

$$k = \frac{C_0 - C(t)}{t} \quad (5)$$

which allows to calculate half-lives $\tau_{1/2}$ (day):

$$\tau_{1/2} = \frac{C_0}{2 \bullet k} \quad (6)$$

If mineralization or biodegradation kinetic model was not specified, first-order kinetics was assumed. In two cases (Lisiecki et al., 2014; Woźniak-Karczewska et al., 2019), mineralization half-lives were estimated using theoretical maximum amount of CO_2 that could be emitted in a given experimental setup as input.

2.2. Statistical uncertainty and analysis of variance (ANOVA)

Statistical uncertainties were calculated as described by Golsteijn et al. (2013). Briefly, we assigned a Student t-distribution with $(n - 1)$ degrees of freedom to the $\log_{10}$-transformed half-life ($\log(\tau_{1/2})$), with the standard error of the mean (*SEM*) values calculated directly from obtained individual experimental points taking into account number of data points. Data were excluded from calculation of *SEM* if less than 3 experimental points were available:

$$SEM_{\log(\tau_{1/2})} = \frac{s}{\sqrt{n}} \quad (7)$$

$$\log(\tau_{1/2}) \sim \overline{log\ \tau_{1/2}} + t_{90} \bullet SEM_{\log(\tau_{1/2})} \quad (8)$$

where $\overline{log\ \tau_{1/2}}$ is the predictive mean of $\log(\tau_{1/2})$, t_{90} is the value of t-distribution with 90% confidence interval, *SEM* is the standard error of the mean, s is the sample standard deviation, and n is the number of data points.

To find statistical significance of the effects of the type of blend, type of experimental system, or degradation endpoint on the half-lives, a two-way analysis of variance (ANOVA) was used. The half-lives were $\log_{10}$-transformed as it improved normality of distribution of the independent variable, as verified by Kolmogorov-Smirnov test. As the design is unbalanced, the Type-III approach to calculate sums of squares was preferred. To identify where significant differences in ($\log_{10}$-transformed) half-lives occur, ANOVA was followed by pairwise post-hoc multiple comparison tests based on the Tukey's Honest Significant Difference (HSD) method. Differences were considered statistically significant if $p < 0.05$. The ANOVA and the Tukey's HSD analyses were performed employing the Anova and TukeyHSD functions available in the R packages car and stats, respectively). R version 4.2.1 was used (R Core Team, Vienna, Austria).

3. Results and discussion

3.1. Evolution of published studies

Fig. 1 shows temporal evolution of studies retrieved from the literature review. It shows that the number of articles published annually rose until ca. 2010, followed by decline (2011–2013) and stabilization (2014–2019). This may reflect responses to policy regulations, as use of biodiesel was first promoted as seemingly more sustainable alternative to petroleum diesel fuel (Guo et al., 2015), leading to series of early EU directives that encouraged production and use of biodiesel as renewable fuel alternative (The European Parliament, 1998a, 1998b, 2003, 2009, 2015, 2018). Increasing awareness about competition with food production and potential negative influence on the environment, lead however to the 2009 RED directive (The European Parliament, 2009) limiting the use of 1st generation biofuels.

3.2. Biodegradation and mineralization half-lives

Table 1 provides an overview of the collected data and resulting log-transformed half-lives ($\log_{10}(\tau_{1/2})$) together with their statistical uncertainties, grouped according to the type of degradation endpoint, blend, experimental system and aeration conditions. All individual data

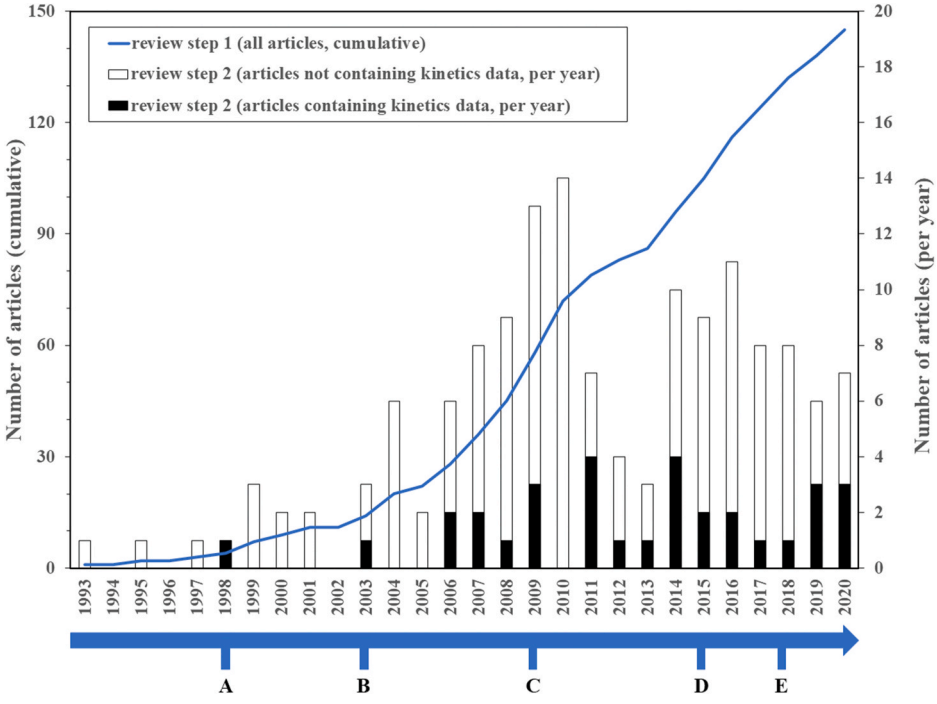

Fig. 1. Temporal evolution of articles and data retrieved from the literature review . **A** – Directive 98/69/EC and Directive 98/70/EC (The European Parliament, 1998a, 1998b) which "sets technical specifications on health and environmental grounds for fuels to be used for vehicles equipped with positive-ignition and compression-ignition engines"; **B** – Directive 2003/30/EC (The European Parliament, 2003) which "aims at promoting the use of biofuels or other renewable fuels to replace diesel or petrol for transport purposes in each Member State, with a view to contributing to objectives such as meeting climate change commitments, environmentally friendly security of supply and promoting renewable energy sources."; **C** – Directive 2009/28/EC (RED) (The European Parliament, 2009) which "establishes a common framework for the promotion of energy from renewable sources (…) and establishes sustainability criteria for biofuels and bioliquids."; **D** - Directive (EU) 2015/1513 (The European Parliament, 2015) which is an amendment to 2009/28 (RED); **E** – Directive (EU) 2018/2001 (RED II) (The European Parliament, 2018) which "establishes a common framework for the promotion of energy from renewable sources (…) and establishes sustainability and greenhouse gas emissions saving criteria for biofuels, bioliquids and biomass fuels.". (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

points are presented in the Appendix A; Table A.1), while Fig. 2 shows a comparison between biodegradation and mineralization half-lives for three selected fuels (diesel fuel, B20 and B100) in aquatic and terrestrial systems, irrespective of aeration conditions.

Table 1 shows that there is approximately 3 times more data points on mineralization kinetics (142 points, including 114 data points for biodiesel fuels) when compared to biodegradation kinetics (56 data points, including 44 data points for biodiesel fuels). The vast majority of data represents aerobic conditions. Studies in aquatic and terrestrial media are approximately equally represented. Across all data points, biodegradation half-lives ranged from 9 to 62 days, which falls within the half-lives range measured for aliphatic hydrocarbons (9–28 days), but can be shorter that half-lives measured for some aromatic hydrocarbons (17–120 days) and some polycyclic aromatic hydrocarbons (PAH) (10–360 days) (Charalampous et al., 2021; Coulon et al., 2010; Heath et al., 1993; Roslund et al., 2018). Across all data points, mineralization half-lives ranged from 16 to 1820 days, which is in range of values reported for various organic compounds (Fantke et al., 2018). Although aerobic mineralization half-lives were ~3 times longer than aerobic mineralization half-lives, a two-way ANOVA performed for terrestrial systems to analyse the effect of degradation endpoint and type of blend on half-lives revealed that none of these two variables had a statistically significant effect on the half-lives ($p = 0.072$ and $p = 0.83$ for degradation endpoint and type of blend, respectively). This lack of statistical significance of the degradation endpoint is in contrast to findings from studies testing mineralization and biodegradation of hydrocarbons and other organic contaminants simultaneously (Atlas and Bartha, 1972; Marchal et al., 2003; Penet et al., 2006; Smith et al., 1997; Solano-Serena et al., 2000; Volke-Sepulveda et al., 2003). For example, degradation mineralization of *n*-paraffins in crude oil was slower than their biodegradation, and attributed this to the formation of intermediates which were inaccessible for microorganisms (Atlas and Bartha, 1972). Similar observations were made for phenanthrene and pyrene (Smith et al., 1997), volatile hydrocarbons of gasoline (Solano-Serena et al., 2000), hexadecane (Volke-Sepulveda et al., 2003), diesel and biodiesel (Marchal et al., 2003; Penet et al., 2006). The lack of

Table 1
Arithmetic mean of $\log_{10}$-transformed biodegradation and mineralization half-lives, log ($\tau_{1/2}$), with τ in days, of diesel and biodiesel fuels and their statistical uncertainty represented by the product of standard error of the mean (SEM) and the value of *t*-distribution with 90% confidence interval (t_{90}). Values in brackets indicate number of experimental data points, while *NA* indicates insufficient number of data points (<3) to calculate the mean. All data are based on the specific 32 studies identified within the scope of the review – details provided in footnotes. Two-way ANOVA did not identify significant effects of the type of blend or experimental system (see details in text in Sections 3.2 and 3.3).

Data set	Diesel fuel	B2	B5	B20	B40	B50	B60	B80	B100
Mineralization									
All data	2.15 ± 0.19 (32)	3.26 ± 0.16 (4)	2.68 ± 0.55 (9)	2.09 ± 0.34 (23)	1.63 ± 0.63 (4)	1.83 ± 0.32 (13)	1.56 ± 0.65 (4)	1.51 ± 0.34 (9)	1.78 ± 0.20 (44)
Aerobic (all systems)	2.15 ± 0.19 (32)	3.26 ± 0.16 (4)	2.68 ± 0.55 (9)	2.09 ± 0.34 (23)	1.63 ± 0.63 (4)	1.83 ± 0.32 (13)	1.56 ± 0.65 (4)	1.51 ± 0.34 (9)	1.78 ± 0.20 (44)
Anaerobic (all systems)	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*
Terrestrial systems	2.25 ± 0.22 (24)	3.26 ± 0.28 (3)	2.78 ± 0.60 (7)	2.33 ± 0.43 (15)	NA	2.18 ± 0.40 (8)	NA	2.13 ± 0.72 (3)	2.13 ± 0.24 (25)
Aerobic conditions[a]	2.25 ± 0.22 (24)	3.26 ± 0.28 (3)	2.78 ± 0.60 (7)	2.33 ± 0.43 (15)	NA	2.18 ± 0.40 (8)	NA	2.13 ± 0.72 (3)	2.13 ± 0.24 (25)
Anaerobic conditions	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*
Aquatic systems	1.85 ± 0.39 (8)	*NA*	*NA*	1.64 ± 0.58 (8)	*NA*	1.28 ± 0.15 (5)	*NA*	1.20 ± 0.18 (6)	1.32 ± 0.26 (19)
Aerobic conditions[b]	1.85 ± 0.39 (8)	*NA*	*NA*	1.64 ± 0.58 (8)	*NA*	1.28 ± 0.15 (5)	*NA*	1.20 ± 0.18 (6)	1.32 ± 0.26 (19)
Anaerobic conditions	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*
Biodegradation									
All data	1.51 ± 0.21 (12)	*NA*	*NA*	1.48 ± 0.34 (8)	*NA*	1.11 ± 0.84 (4)	*NA*	*NA*	1.35 ± 0.23 (26)
Aerobic (all systems)	1.55 ± 0.29 (9)	*NA*	*NA*	1.50 ± 0.40 (7)	*NA*	1.27 ± 1.31 (3)	*NA*	*NA*	1.38 ± 0.23 (20)
Anaerobic (all systems)	1.40 ± 0.35 (3)	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	1.24 ± 0.51 (6)
Terrestrial systems	1.71 ± 0.15 (8)	*NA*	*NA*	1.80 ± 0.14 (5)	*NA*	*NA*	*NA*	*NA*	1.63 ± 0.12 (14)
Aerobic conditions[c]	1.75 ± 0.15 (7)	*NA*	*NA*	1.80 ± 0.14 (5)	*NA*	*NA*	*NA*	*NA*	1.61 ± 0.13 (13)
Anaerobic conditions[d]	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*
Aquatic systems	1.11 ± 0.49 (4)	*NA*	*NA*	0.94 ± 0.66 (3)	*NA*	*NA*	*NA*	*NA*	1.02 ± 0.36 (12)
Aerobic conditions[e]	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	0.94 ± 0.58 (7)
Anaerobic conditions[f]	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	*NA*	1.13 ± 0.60 (5)

[a] 89 data points from references (Cruz et al., 2020; Dos Anjos et al., 2018; Horel and Schiewer, 2011, 2014, 2016, 2020; Lapinskiene et al., 2006; Lisiecki et al., 2014; Meyer et al., 2014; Pinto Mariano et al., 2008; Schiewer and Horel, 2017; Silva et al., 2012; Soares Junior et al., 2009; Woźniak-Karczewska et al., 2019).
[b] 53 data points from refs (Demirbaş, 2009; Montagnolli et al., 2019; Ng et al., 2015; Pasqualino et al., 2006; Pinto Mariano et al., 2008; Vauhkonen et al., 2011; Yassine et al., 2013; Zhang et al., 1998).
[c] 29 data points from refs (Chen et al., 2019; Lapinskiene et al., 2006; Li et al., 2016b; Thomé et al., 2014; Wieczorek et al., 2015).
[d] 2 data points from ref (Lapinskiene and Martinkus, 2007) (insufficient number of data points to calculate the mean).
[e] 16 data points from refs (Cyplik et al., 2011; DeMello et al., 2007; Owsianiak et al., 2009; Zhang et al., 1998).
[f] 9 data points from refs (Corseuil et al., 2011; Cyplik et al., 2011; Makareviciene and Janulis, 2003).

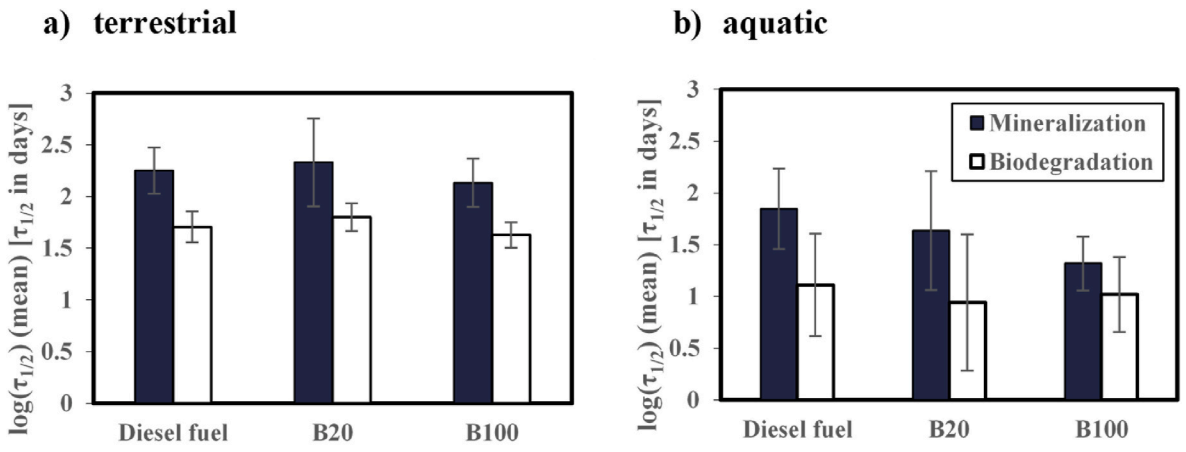

Fig. 2. Arithmetic mean of $\log_{10}$-transformed biodegradation and mineralization half-lives, log ($\tau_{1/2}$), with τ in days, of diesel, B20 and B100 fuels in **(a)** terrestrial systems (aerobic and anaerobic data combined) and **(b)** aquatic systems (again, aerobic and anaerobic data combined). Error bars indicate $SEM_{\log(\tau_{1/2})} \bullet t_{90}$ values. Presented data were derived from the data shown in Table 1. Two-way ANOVA revealed that the type of experimental systems did have a statistically significant effect on biodegradation half-lives ($p = 0.03$), while the type of blend did not ($p = 0.87$). Tukey's HSD test found that the mean value of biodegradation half-life was significantly different between terrestrial and aquatic systems for pairs B0–B100, B20–100 and B100–B100. Two-way ANOVA also revealed that the type of degradation endpoint did have a statistically significant effect on half-lives ($p = 0.03$), and Tukey's HSD test found that the mean value of biodegradation half-life was significantly different between mineralization and biodegradation endpoints for pairs B0–B100 and B20–B100. Results of these ANOVA runs must be treated with care as more detailed analysis of data presented in Table 1, performed for anaerobic data only, did not confirm statistically significant effects of the type of experimental system or degradation endpoint.

statistically significant effect in our study must therefore be due to averaging effect, where data from different (unbalanced) experiments are combined. Several factors can explain the difference between mineralization and biodegradation kinetics. First, the uptake of the substance by the cell and the release of carbon dioxide, a multi-step oxidation must take place. This process incorporates different enzymes, which is why the release of CO_2 is delayed when compared to the disappearance of parent compound from the system. Second, microbial activity is associated with the formation of cellular biomass. Thus, part of the removed carbon source is bound in the form of new biomass, which can be further mineralised by other microorganisms only after cell lysis. This might also have an impact on degradation kinetics, since fatty acids are integral part of phospholipids which form microbial membranes, and hence their transformation into CO_2 will be delayed. Third, during the oxidation stage branch and cyclic hydrocarbons originating from diesel fuel in a blend, intermediates which do not undergo fast oxidation may be formed, or due to their presence the process is delayed (Horel and Schiewer, 2009, 2011; Lapinskiene et al., 2006; Marchal et al., 2003; Penet et al., 2006; Van De Steene and Verplancke, 2007; Zhang et al., 1998).

Collected data for biodegradation and mineralization in anaerobic conditions were too scarce (a sum of 11 experimental points) to allow for a robust comparison with aerobic conditions. Individual studies suggest that biodegradation under aerobic conditions (for B100, B20 and diesel fuel) was faster than in anaerobic conditions (Bregnard et al., 1996; Cason et al., 2019; Cyplik et al., 2011; Queiroz et al., 2016; Salminen et al., 2004; Varrone et al., 2017). The vast majority of tests (*i.e.*, a sum of 187 experimental points) was performed in aerobic conditions, which may be explained by the fact that aerobic conditions are generally easier to control in laboratory conditions. Anaerobic-oriented studies focus on fuel stability and tank corrosion during storage, which were outside the scope of this review (Gupta, 2020; Leung et al., 2006; Luz et al., 2018).

3.3. Differences between fuels and experimental systems

Data presented in Table 1 and Fig. 2 show that petroleum diesel fuel had longer ($\log_{10}$-transformed) half-lives than biodiesel (B100) (~1.5 times for biodegradation and ~2.5 times for mineralization). However, Table 1 also shows that blends B2 and B5 in all considered systems had longer half-lives when compared to those petroleum diesel fuel. This is unexpected, because mineralization rates generally increase with increasing biodiesel content (Chen et al., 2019; Soares Junior et al., 2009). Indeed, two-way ANOVA did not identify significant effect of the type of blend ($p = 0.28$), which we attribute to relatively low number of experimental data points for lower blends and averaging across various which masks differences in mineralization and biodegradation half-lives between different fuels.

Biodiesel is inherently more biodegradable to microorganisms than petroleum diesel fuel, mainly because biodegradation of biodiesel uses the same set of enzymes as in fat decomposition and these enzymes are generally abundant (Aktas et al., 2010; Lapinskiene and Martinkus, 2007; Silva et al., 2012; Zhang et al., 1998). In the first stage of biodegradation, extracellular hydrolysis of the ester takes place with the participation of hydrolases, and then the resulting fatty acid undergoes β-oxidation catalysed by Acetyl-CoA dehydrogenase (Aktas et al., 2010; Fang and McCormick, 2006; Lapinskiene and Martinkus, 2007; Zhang et al., 1998) or is directly incorporated into cell membrane structures *e.g.* phospholipids, which is energetically preferential from the cellular point of view (Von Wallbrunn et al., 2003). Diesel fuel, on the other hand, is a mixture of thousands of compounds with different structures, consisting mainly of aliphatic and aromatic hydrocarbons, which means that a wider range of specific enzymes is needed to biodegrade it (Zhang et al., 1998). In the case of aliphatic hydrocarbons, their oxidation process results in formation of fatty alcohols, and further fatty acids. However, the initial energy input, as well as structure of hydrocarbon, determines the final energy gain, making linear alkanes much more attractive energy source than branch alkanes and especially than cycloalkanes (Ławniczak et al., 2020). In regard to benzene, its derivatives or polycyclic aromatic hydrocarbons, the degradation pathways are even more complicated and require both different specific enzymes and higher energy input. Hence, shorter mineralization half-lives of biodiesel are indicative of the lower amount of intermediates, biomass or non-degradable compounds formed, which may be due to the simpler structure and shorter metabolic pathway than for *e.g.* aromatic compounds present in diesel oil.

The terrestrial data include various agricultural soils and natural

soils, artificial porous matrices with varying moisture content, composition and degrading microorganisms. The aqueous systems include both artificial mineral media and natural waters of varying compositions and types of microorganism. When averaging half-lives across those data we observed generally longer half-lives in terrestrial systems (2–8 times longer depending on the blend) when compared to aquatic systems, but two-way ANOVA did not confirm significant effect of the type of experimental systems ($p = 0.15$). Pinto Mariano et al. (2008) showed that mineralization kinetics of biodiesel blends in aqueous matrix was ~2 times faster than in soils. This can be explained by the fact that soil is much more complicated matrix than aqueous media, influencing accessibility and bioavailability of carbon to degrading organisms (Ledezma-Villanueva et al., 2016; Reichenberg and Mayer, 2006). Indeed, in sorption of contaminants to soil constituents like clay or organic carbon can influence and their transport rate are strongly influenced by soil porosity, the presence of organic matter and water content (Gupta, 2020; Ledezma-Villanueva et al., 2016). The presence of organic matter can additionally lead to biological activity that competes with the biodegradation of the xenobiotic, which, given the limited availability of micro- and macroelements and electron acceptors, can significantly affect the observed biodegradation (Diplock et al., 2009).

3.4. Limitations and data gaps

The largest limitation of the present study is the unbalanced design of data sets for statistical analysis, with different number of data points representing a wide range of experimental setups. Lack of statistically significant effect of the type of fuel can be attributed to relatively low number of experimental data points for lower blends and uncertainties which masked differences in mineralization and biodegradation half-lives between different fuels. Indeed, the vast majority of studies included in this review showed a significant effect of the type of blend, with higher biodegradation kinetics of blends with higher biodiesel content (Table A.1 and references therein).

Next to differences in numbers of data, factors other than type of biodiesel fuel and experimental system could influence biodegradation and mineralization kinetics of biodiesel fuels. They include: origin and composition of the biodiesel, temperature in which kinetics was measured, or total content of fuel in the experimental media. Due to limited availability of data their consideration in the statistical analysis proved challenging. Yet we elucidated the role of these factors by investigating selected studies form the literature review. Thereby, we found that the origin of biodiesel fuel (*i.e.,* rapeseed methyl ester, rapeseed ethyl ester or soy methyl ester) is not expected to be an important factor for biodegradation. Others already showed that differences in biodegradation kinetics between biodiesels of different origins were very small and statistically insignificant (Demirbaş, 2009; Makareviciene and Janulis, 2003; Ng et al., 2015; Zhang et al., 1998). We also found that the influence of the amount of fuel in the experimental medium on biodegradation and mineralization kinetics is not obvious. Analysis of data from individual studies shows that higher concentration of fuel introduced generally correlates with slower biodegradation and mineralization kinetics, although these observations are based on 3 to 6 data points per study and therefore not straightforward to interpret (Lapinskiene et al., 2006; Li et al., 2016b; Wieczorek et al., 2015; Zhang et al., 1998). This data gap should be investigated in future experimental studies. Finally, we found that temperature can be a factor influencing biodegradation rates. Horel and Schiewer (2011) compared biodegradation kinetics at low temperatures (6 °C) with that in higher ranges of values (up to 20 °C), showing significantly slower mineralization at 6 °C. Inclusion of their data point in our study may therefore lead to bias for B20 and B100 blends. For all other data points included in our review, temperature was in range of 20–30 °C and roughly equally spread across blends and experimental systems, suggesting that its influence on statistical analysis was relatively small.

4. Practical implications

Reviewed data indicate that biodiesel fuels in aerobic aquatic and terrestrial systems is expected to be degraded at rates which are generally shorter than those of petroleum hydrocarbons. This may suggest that bioremediation of biodiesel fuels may not necessarily require the same approaches as that of diesel fuel. Thus, employing more active remediation techniques, like addition of surfactants to increase bioavailability or augmentation with degrading microorganisms may not necessarily bring more benefits than what can be gained though natural attenuation mechanisms. Fernández-Álvarez et al. (2007) already showed that bioaugmentation did not significantly speed up biodegradation of aliphatic and aliphatic hydrocarbons in the presence of biodiesel in marine ecosystems.

In environmental risk assessment (ERA), environmental impact assessment (EIA) and life cycle impact assessment (LCIA), assessing environmental fate of hydrocarbons in soils often relies on mineralization rate constants derived from aquatic biodegradation data using constant extrapolation factors (*e.g.*, assuming that first-order rate constant in soil is 2 times lower when compared to that in the freshwater environment) (Bamard et al., 2011; Fantke et al., 2018). Our kinetic data can be used to better assess environmental fate of biodiesel fuels in soils by using first-order mineralization rate constant derived from measured data in terrestrial environments, rather than using extrapolated values. Our kinetic data can also be used in cases where aquatic mineralization rate constants is estimated using predictive biodegradation models. For application in assessments where site conditions (be it soil or aquatic system) are not known, like in generic LCIA, we recommend using average first-order mineralization rate constants derived from kinetic data reported in Table 1. Given large variability in measured mineralization half-lives reported in Table 1, average kinetic parameters may, however, not necessarily apply for application in regionalized assessments, including ERA and EIA. In those cases, we recommend assessors to estimate rate contents at the site. Alternatively, our data reported for those aquatic and terrestrial systems which best match conditions at the site (Table A.1), could be used.

5. Conclusions

We showed that (1) mineralization half-lives are from 2.5 to 5 times longer than biodegradation half-lives; (2) the half-lives of pure biodiesel (B100) from 1.2 to 3.4 times shorter than those of petroleum diesel fuel; and (3) the half-lives are generally longer (from 2.5 to 8.5 times) in terrestrial systems when compared to aquatic systems. Lack of statistically significant differences in biodegradation and mineralization half-lives between blends can be attributed to unbalanced design and masking effect when uncertainties are considered. This relatively large variability in biodegradation and mineralization half-lives stresses the need for site-specific evaluation of kinetic parameters. The half-lives reported in the present study can be, however, used for assessment of environmental fate and resulting risks and impacts in conditions where site parameters are not known. Generally higher biodegradability of biodiesel blends when compared to petroleum diesel fuel suggest that passive approaches to degrade and dissipate biodiesel contaminants in situ, like monitored natural attenuation, may be relevant strategies for biodiesel fuels.

Credit author statement

Wiktoria Wilms: Formal analysis, Investigation, Data Curation, Writing - Original draft preparation; **Jan Homa**: Investigation, Writing - Original draft preparation; **Marta Woźniak-Karczewska**: Visualization, Writing- Reviewing and Editing; **Mikołaj Owsianiak:** Conceptualization, Methodology, Supervision, Writing- Reviewing and Editing; **Łukasz Chrzanowski**: Conceptualization, Methodology, Supervision, Writing- Reviewing and Editing, Project Administration; Funding

acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

I have attached the data in the Attach file step (Appendix A)

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.137236.

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