



## Review

# Anaerobic biodegradation of (emerging) organic contaminants in the aquatic environment



Ann-Kathrin Ghattas<sup>1</sup>, Ferdinand Fischer<sup>1</sup>, Arne Wick, Thomas A. Ternes\*

Federal Institute of Hydrology (BfG), D-56068 Koblenz, Am Mainzer Tor 1, Germany

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## ABSTRACT

Although strictly anaerobic conditions prevail in several environmental compartments, up to now, biodegradation studies with emerging organic contaminants (EOCs), such as pharmaceuticals and personal care products, have mainly focused on aerobic conditions. One of the reasons probably is the assumption that the aerobic degradation is more energetically favorable than degradation under strictly anaerobic conditions. Certain aerobically recalcitrant contaminants, however, are biodegraded under strictly anaerobic conditions and little is known about the organisms and enzymatic processes involved in their degradation. This review provides a comprehensive survey of characteristic anaerobic biotransformation reactions for a variety of well-studied, structurally rather simple contaminants (SMOCs) bearing one or a few different functional groups/structural moieties. Furthermore it summarizes anaerobic degradation studies of more complex contaminants with several functional groups (CMCs), in soil, sediment and wastewater treatment. While strictly anaerobic conditions are able to promote the transformation of several aerobically persistent contaminants, the variety of observed reactions is limited, with reductive dehalogenations and the cleavage of ether bonds being the most prevalent. Thus, it becomes clear that the transferability of degradation mechanisms deduced from culture studies of SMOCs to predict the degradation of CMCs, such as EOCs, in environmental matrices is hampered due the more complex chemical structure bearing different functional groups, different environmental conditions (e.g. matrix, redox, pH), the microbial community (e.g. adaptation, competition) and the low concentrations typical for EOCs.

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\* Corresponding author.

E-mail address: [ternes@bafg.de](mailto:ternes@bafg.de) (T.A. Ternes).

<sup>1</sup> Ann-Kathrin Ghattas and Ferdinand Fischer contributed equally to this work.

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## 1. Introduction

As reflected by the vast knowledge about aerobic degradation mechanisms in comparison to anaerobic ones (e.g. Neilson and Allard, 2008), aerobic conditions are generally reported to be more effective for the removal of most organic contaminants. Several pollutants, such as highly halogenated aromatic compounds, however, have been shown to be more easily degraded under strictly anaerobic conditions (Vogel et al., 1987). However, little is known about the organisms and enzymatic processes involved in anaerobic degradation of these compounds. Anaerobic conditions are frequently occurring in environmental matrices, such as soils, sediments and aquifers (Bethke et al., 2011; Champ et al., 1979). The thermodynamic ladder of geomicrobiology suggests stratification into a series of redox zones in which biological respiration progressively yields less energy according to the half-potentials of the available corresponding electron acceptors (Bethke et al., 2011; Champ et al., 1979). It is however known today, that this stratification is not always quite as clear cut as previously assumed (e.g. Appelo and Postma, 2005; Huettel et al., 2003). Nevertheless, the types of microorganisms and metabolisms adapted towards these oxygen-free habitats strongly depend on the availability of electron acceptors and their respective energy yield.

After oxygen depletion by aerobic respiration, nitrate can be used as alternative electron acceptor by **nitrate reducing** or denitrifying bacteria. As denitrifying bacteria are commonly facultative anaerobes which easily switch between aerobic and anoxic respiration, it is difficult to distinguish between an aerobic and anoxic reaction under denitrifying conditions, if the exclusion of oxygen cannot be guaranteed (Körner and Zumft, 1989; Meiberg et al., 1980; Wilson and Bouwer, 1997). Due to the potential of aerobic degradation under denitrifying conditions, these are not considered in this review and the term “anaerobic” will be used for conditions/reactions excluding oxygen and nitrate as electron acceptors.

**Manganese and iron reduction** takes place under redox potentials below those for nitrate reduction. Under these conditions, contaminants, such as benzene, toluene, ethylbenzene and xylene (BTEX) (Abu Laban et al., 2010; Anderson et al., 1998; Jahn et al., 2005; Kunapuli et al., 2008; Rooney-Varga et al., 1999; Villatoro-Monzon et al., 2003, 2008; Weelink et al., 2010), halogenated aromatics (Kazumi et al., 1995), and carbon tetrachloride (Boopathy, 2002) are known to be degraded. Furthermore, abiotic degradation by iron(II) produced by microbial iron(III) reduction has been reported for nitro aromatic compounds (Hofstetter et al., 1999; Tor et al., 2000), dichlorodiphenyltrichloroethane (DDT) (Li et al., 2010) and for the antibiotic sulfamethoxazole (SMX) (Mohatt et al., 2011).

At redox conditions below the iron reducing zone, dissolved

sulphate anions are used as alternative electron acceptor. The relevance of **sulphate reducing** microorganisms for the degradation of emerging organic contaminants (EOCs) derives from the wide range of compounds serving as electron donors (Barton and Fauque, 2009; Liamleam and Annachhatre, 2007). Among these are monoaromatic compounds such as BTEX (Abu Laban et al., 2009; Lovley et al., 1995; Musat and Widdel, 2008) as well as polycyclic aromatic substances like naphthalene or anthracene (Bergmann et al., 2011; Coates et al., 1996a; Galushko et al., 1999; Meckenstock et al., 2000; Ramsay et al., 2003), alkanes (Kniemeyer et al., 2007; So and Young, 1999; Widdel and Rabus, 2001) and chlorinated compounds (Boopathy, 2002; Drzyzga et al., 2001; Savage et al., 2010; van Eekert and Schraa, 2001).

In the absence of inorganic electron acceptors except CO<sub>2</sub>, **methanogenesis** plays an important role for the biodegradation of organic matter. Among others, benzene (Kazumi et al., 1997; Ulrich et al., 2005; Weiner and Lovley, 1998), toluene (Grbić-Galić and Vogel, 1987; Meckenstock, 1999), carbon tetrachloride (Boopathy, 2002), 2,4,5-trichlorophenoxyacetic acid (Gibson and Sufita, 1990), phenoxyethanol (Frings and Schink, 1994), dinoseb (Hammill and Crawford, 1996) halogenated compounds with a methyl or ethyl moiety as carbon backbone (Belay and Daniels, 1987; Bouwer and McCarty, 1983), halogenated aromatic compounds (Dolfing and Tiedje, 1991; Häggblom et al., 2000; Liang et al., 2013; Stams et al., 2006), 1,1,2,2-tetrachloroethane (Lorah and Voytek, 2004), the herbicide picloram (Ramanand et al., 1993) and the antimicrobial agent triclosan (Veetil et al., 2012) have been reported to be degraded under methanogenic conditions. However, methanogens cannot directly degrade these complex molecules and thus depend on the supply of electron donors and substrate (H<sub>2</sub>/CO<sub>2</sub>, formate/acetate) by other microorganisms. Therefore, methanogens usually co-exist in a microbial consortium with hydrolytic, fermentative syntrophic and acetogenic microbes (Madigan et al., 2000; Nazaries et al., 2013; Stams et al., 2005). **Fermenting** organisms can also use a large variety of organic substrates, including aromatic compounds, such as dinoseb (Hammill and Crawford, 1996) or phenoxyethanol (Frings and Schink, 1994), as electron acceptor. Heterotrophic **acetogenic** bacteria, which oxidize reduced organic fermentation products (e.g. short chain volatile acids, ketones or alcohols) to substrates for methanogenesis (i.e. acetate and H<sub>2</sub>), are part of the consortium, if hydrogen is effectively removed by the methanogens (Bryant et al., 1967, 1977; Stams, 1994; Stams et al., 2005). Under high hydrogen concentrations, this link between fermentation and methanogenesis can be provided by sulphate reducing organisms, which oxidize reduced organic fermentation products. This link is, however, not always advantageous, as sulphate reducers partially compete for substrates, such as hydrogen and acetate, with

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