



Review

Perfluorooctane sulphonate (PFOS) throughout the food production chain

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ABSTRACT

Perfluorooctane sulphonate (PFOS) is a persistent organic pollutant with adverse effects on human health. Since dietary intake plays an important role in human exposure, the transfer of PFOS throughout the food chain needs further investigation. The aim of this paper is to give an overview of PFOS concentrations and transfer for the various chain steps from farm-to-fork. This reveals that most research focused on levels of PFOS in surface water and fish but data on soil and crops are largely missing. Furthermore, the uptake of PFOS by farm animals and subsequent transfer into meat and animal products needs further attention, as these products will eventually be consumed by the human population. Once the necessary data gaps are filled, the contribution of the various chain steps on the total PFOS intake can be established. Moreover, the effect of pollution events on the food chain can be established enabling appropriate actions in order to protect consumer health.

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Contents

1. Introduction	1
2. PFOS in water	2
3. PFOS in soil	3
4. PFOS in crops	3
5. PFOS in animals	3
6. PFOS in food	4
7. Conclusions	5
Acknowledgements	5
References	5

1. Introduction

Perfluorinated compounds (PFCs) are compounds in which all hydrogen atoms attached to carbon atoms are replaced with fluorine atoms. They are persistent chemicals that are widely distributed in the environment. Exposure to PFCs has resulted in hepatotoxicity, developmental toxicity, immunotoxicity, adverse hormonal effects and carcinogenic potency in animal studies (Clarke et al., 2010; Hölzer et al., 2008; Wilhelm, Kraft, Rauchfuss, & Hölzer, 2008). The group of PFCs includes perfluorooctane sulphonate (PFOS), which is the most frequently detected compound in food products with generally the highest concentrations within the PFC group (FSA, 2009). PFCs can be found in surface water,

sewage sludge, soil, sediment and air across the world, with elevated concentrations in relatively populated and industrialised regions, especially near production sites (Giesy & Kannan, 2001; Houde, Martin, Letcher, Solomon, & Muir, 2006; Lau et al., 2007; OECD, 2002). They can also be found in remote regions such as the Arctic (Dietz, Bossi, Rigét, Sonne, & Born, 2008; Giesy & Kannan, 2002). At the moment, there is a lack of information on the exact sources of PFCs in the environment. Contamination in wildlife varies among species and locations which indicates multiple emission sources. PFCs may be released into the environment by manufacturing or disposal operations or during the useful lifetime of a product (Houde et al., 2006). Several sources, such as discharge of industrial and municipal wastewater, fire-fighting operations at military bases and airports, and landfill leachate, may be responsible for elevated exposure to PFCs in urban areas (Houde et al., 2006; Vestergren & Cousins, 2009). In 2006, high concentrations of PFCs

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were detected in surface and drinking water in Germany (Skutlarek, Exner, & Farber, 2006). As a result of this, an extensive monitoring program was initiated analysing soil, surface and drinking water to identify sources of PFC exposure (LANUV, 2008). This monitoring program showed that the source of the PFC contamination in this case was the widespread use of soil conditioner, which had been mingled with industrial waste. This contamination has led to elevated PFC concentrations in the blood plasma of children and adults living in the area (Hölzer et al., 2008). A similar pollution event occurred in Alabama, resulting in increased PFOS concentrations in soil and sludge (EPA, 2009; Renner, 2009).

Aqueous fire fighting foams (AFFFs) presumably are the most prominent source of widespread environmental dispersal of PFOS. As a result of this, PFOS has increased in a range of wildlife over the period 1969–2002 (Paul, Jones, & Sweetman, 2009). The 3M company, the dominant global producer of PFOS responsible for the large majority of total global production volumes, phased out production in 2002. Since then, the first declines in human exposure have been reported (Brooke, Footitt, & Nwaogu, 2004; Clarke et al., 2010). In the EU, a restriction is laid down on the marketing and use of PFOS following directive 2006/122/EEC as an amendment of directive 76/769/EEC. This restriction covers all products to which PFOS is added intentionally (e.g. textiles). Moreover, the use of PFOS in the plating industry should be minimised and the use of existing stocks of fire-fighting foams containing PFOS is allowed until June 2011. On-going uses in the aviation industry (hydraulic fluids), the semiconductor industry and the photographic industry (coatings) are not supposed to pose a relevant risk to the environment or human health if releases into the environment and workplace exposure are minimised. Recently, the EU commission has recommended the monitoring of PFOS and PFOA in food (2010/161/EU). There is, however, currently no legislation for PFOS in food and feed within the EU. Their use in plastics and coatings for food contact materials has been approved in the Netherlands and Germany (EFSA, 2008). However, as soon as safer alternatives are feasible, the use of PFOS is phased out. Recently, PFOS has been designated as a Persistent Organic Pollutant under the Stockholm Convention (United Nations Environmental Programme, 2009). PFOS is also added to the OSPAR list, an international governmental cooperation for the protection of the Marine environment of the North-East Atlantic, list of Chemicals for Priority Action in 2003 (OSPAR, 2006).

PFOS emissions are estimated to continue over the next decade from stain-resistant carpets and from PFOS containing AFFFs until the latter is banned in Europe in 2011 (Halldorsson et al., 2008). Since PFOS has an estimated half-life of 41 years in the environment (Clarke et al., 2010), it is likely to be of continued public health interest. The route of human exposure to PFOS, however, has not been well characterised (Halldorsson et al., 2008). Although non-food sources such as house dust and outdoor air are seen as possible contamination routes, they contribute less than 2% of the average intake (EFSA, 2008). Dietary intake is, therefore, considered as an important source of exposure to PFOS

(Ericson, Martí-Cid, et al., 2008; Fromme, Tittlemier, Völkel, Wilhelm, & Twardella, 2009; Tittlemier et al., 2007; Trudel et al., 2008; Wilhelm et al., 2008). However, the contamination route from farm-to-fork is largely unknown (Fig. 1). It is, therefore, necessary to gain insight into the levels and transfer of PFOS for the various steps in the food supply chain. Once the transfer of PFOS through the food chain can be quantified, the relevance of the various chain steps for human intake can be established. The aim of this paper is, therefore, to give an overview of PFOS levels as published in literature for the various steps in the food supply chain together with information available on the transfer of PFOS from one step to the next.

2. PFOS in water

PFOS can enter the aquatic environment through manufacturing companies and other sources, which eventually may lead to polluted tap water (Suja, Pramanik, & Zain, 2009). Several review papers have published levels of PFOS in surface water (Chen et al., 2009; Ericson, Nadal, Van Bavel, Lindström, & Domingo, 2008; Fujii, Polprasert, Tanaka, Lien, & Qiu, 2007; Suja et al., 2009). The majority of surface water data originate in Japan and the USA. Although PFOS can be removed from drinking water with activated coal, this treatment is not common. Therefore, monitoring of surface water is relevant to evaluate possible contamination through drinking water (Nakayama, Strynar, Reiner, Delinsky, & Lindstrom, 2010). Direct tap water data are encountered less frequently. An overview of PFOS levels in surface and drinking water can be found in, EFSA (2008). Background levels in drinking water ranged from <0.01 to 50.9 ng/l (with median values between 1 and 5 ng/l) and in surface water from <0.01 to 135 ng/l (with median values around 1 ng/l) (EFSA, 2008). PFOS concentrations can be much higher close to a local contamination source. Point sources were, e.g. identified by Saito et al. (2004), who traced down high levels of PFOS in the Yodo river (up to 526 ng/l) to the Osaka international airport; presumably due to the use of AFFF. Another local source was established in Germany where contaminated soil conditioner was used leading to high PFOS levels in the Moehne river (up to 5900 ng/l) (Skutlarek et al., 2006). Wastewater treatment plants (WWTP) are also often linked to high local concentrations of PFOS (Nakayama et al., 2010). Despite the phasing out of PFOS production in 2002, products containing PFOS are still being used and released into WWTP (Loganathan, Sajwan, Sinclair, Senthil Kumar, & Kannan, 2007). The levels depend upon the fraction of industrial contribution to the WWTP (Becker, Gerstmann, & Frank, 2008; Sinclair & Kannan, 2006; Yu, Hu, Tanaka, & Fujii, 2009) with current maximum levels in the order of 100 µg/kg dry weight (3M, 2001a; Becker et al., 2008; Bossi, Strand, Sortkjær, & Larsen, 2008; Loganathan et al., 2007; Schultz et al., 2006; Senthilkumar, Ohi, Sajwan, Takasuga, & Kannan, 2007; Sinclair & Kannan, 2006; Yu et al., 2009). Mass flow studies in waste water treatment have shown an increase in PFOS from influent to effluent that may be due to biodegradation of precursor compounds during activated sludge treatment

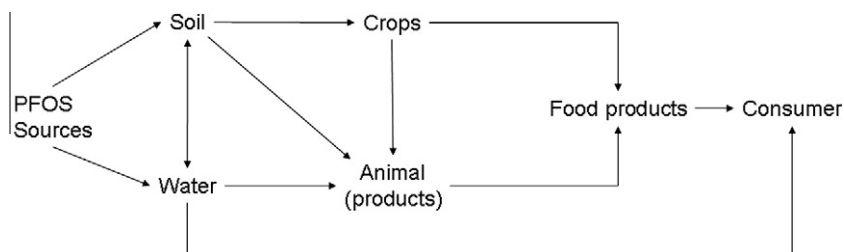


Fig. 1. PFOS transfer throughout the food chain.

(Loganathan et al., 2007; Schultz et al., 2006; Sinclair & Kannan, 2006).

Since there is a lack of representative data for drinking water, EFSA used fresh water as a possible precursor of drinking water for their dietary intake study (EFSA, 2008). A Japanese study showed that there is, indeed, a good correlation between PFOS levels in drinking water and surrounding surface water in a 1:1 ratio (Fujii et al., 2007). This is confirmed by measurements of drinking water produced from highly contaminated surface water (3M, 2001a; Saito et al., 2004; Skutlarek et al., 2006). Although drinking water, in general, is estimated to be a negligible source of PFOS intake (EFSA, 2008; Ericson, Nadal, et al., 2008; Tittlemier et al., 2007), it may become relevant in case of local pollution events (Halldorsson et al., 2008; Hölzer et al., 2008; Hölzer et al., 2009; Vestergren & Cousins, 2009).

3. PFOS in soil

In the German pollution case, the use of waste in a soil conditioner has polluted agricultural soil in various German states. In North Rhine Westphalia this resulted in a temporary soil monitoring program including the analyses of 916 field samples which were suspected of pollution. Despite the obvious emission, levels of the sum of PFOA and PFOS remained below detection in more than half of the samples (57%). A third of the samples contained levels above the detection limit but remained below 100 µg/kg, whereas 8% of the samples had levels between 100 and 500 µg/kg. The highest levels (>1500 µg/kg) were found on two farms which subsequently have been sanitized (LANUV, 2010). Samples from a non-contaminated area in Germany (Bayern) showed that all soil samples ($n = 199$) contained PFOS at levels below 10 µg/kg (LfU, 2007). Additional soil data of PFOS in reviewed literature are, however, lacking.

A number of studies have measured both sediment or sludge and the surrounding surface water (3M, 2001a; Becker et al., 2008; Bossi et al., 2008; Loganathan et al., 2007; Schultz et al., 2006; Senthilkumar et al., 2007; Sinclair & Kannan, 2006; Yu et al., 2009). PFOS in samples from sludge range between <0.2 and 3120 µg/kg (median is 124 µg/kg), with highest levels near a fluorochemical manufacturer site (3M, 2001a). As sludge can be recycled as fertilizers to the soil, it can be a source of PFOS contamination of the soil.

In order to quantify the transfer of PFOS from soil to surface water, it is necessary to obtain data from the same locations. In both the German and Alabama pollution case, samples were taken from surface water, drinking water and soil from the contaminated region (EPA, 2010; LANUV, 2008). Such data can be used to assess the fate of PFOS from soil to surface and drinking water. Apart from field studies, data from experiments establishing the adsorption of PFOS to various soils (Higgins & Luthy, 2006; Johnson, Anschutz, Smolen, Simcik, & Penn, 2007) can be used to estimate the transfer of PFOS from surface water to the soil.

4. PFOS in crops

Measurements in crops are hardly available in peer reviewed literature. Based on the German pollution case, the Ministry of the Environment of North Rhine-Westphalia evaluated the PFOS levels in various crops grown in the contaminated Sauerland region. Maize, grass, rapeseed, black salsify and wheat were sampled in 2006 and 2007. PFOS levels in almost all samples remained below the LOD (1 µg/kg), except for wheat which contained 3 µg/kg in one sample (out of 6 samples) and maize where 38% of the samples ($n = 16$) contained levels above LOD, ranging between 3 and 164 µg/kg (LANUV, 2008). Contaminated soil from the same region

was also used to perform crop growth experiments showing a direct relationship between PFOS levels in plants and soil. Non-contaminated soil, moderately contaminated soil (levels around 300 µg/kg) and highly contaminated soil (levels around 2000 µg/kg) were used to grow maize, wheat, potatoes and grass. Based on these experiments, transfer coefficients for the various crops could be established. Highest transfer coefficients were found for grass (0.078–0.255) whereas wheat showed the lowest transfer coefficient (0.001–0.004) (IME, 2009, 2008). Brooke et al. (2004) found that concentrations in plant vegetative tissue were generally 1–2 times higher than the concentration in the soil. The plants and PFOS concentrations in the soil used in their experiments were, however, not specified. Stahl et al. (2009) found a log-linear relationship between PFOS in soil and plant (wheat, oat, maize, potatoes and ryegrass). The coefficients of the relationship varied for the various crops (Stahl et al., 2009). The transfer from soil to crop, therefore, depends on the concentrations present in the soil, but also on the crop itself. The effect of soil characteristics on PFOS transfer to the crop has not been studied yet.

5. PFOS in animals

PFOS levels are studied much more often in marine animals than in terrestrial animals (Houde et al., 2006). In general, the highest concentrations have been found in the livers of fish-eating animals living close to industrialised areas (Lau et al., 2007). One study reported extremely high liver concentrations of PFOS (between 0.47 and 178.55 µg/g) for a population of field mice living near a fluorochemical plant in Belgium (Hoff et al., 2004). Giesy, Kannan and co-workers have measured PFOS levels in various wildlife (Giesy & Kannan, 2001, 2002; Kannan et al., 2005) showing that PFOS biomagnifies in higher trophic levels (Kannan et al., 2005). In Germany, more than 2000 wild boars have been evaluated for PFOS presence. Almost all samples were positive with levels between 5 and 1592 µg/kg in liver tissue (BfR, 2009; Hütteroth, 2009; LANUV, 2008; LHL, 2007). Meat samples were positive in 40% of the cases, with levels up to 29 µg/kg (LHL, 2007).

Simultaneously measured concentrations in surface water and fish enables the establishment of field-based bioconcentration factors (BCFs) (Houde et al., 2006), i.e. the concentration of PFOS in the fish divided by the concentration in the water. An overview of reported BCFs is given in Table 1. The reported BCF values show a large variation, which may be explained by several factors. First of all, PFOS is known to accumulate in kidney and liver, with liver concentrations several times higher than serum concentrations (Lau et al., 2007). Measurements in edible tissue will thus give other results than measurements in livers and kidneys. Furthermore, the BCF may have been influenced by the presence of precursors that are not detected in the surface water, but may have been metabolized to PFOS in the fish liver (Houde et al., 2006). Finally, local contamination may influence the BCF values obtained. For example, the upper value obtained by Moody, Martin, Kwan, Muir, and Mabury (2002) (log BCF = 5.1) was caused by PFOS measurements downstream of an international airport, presumably caused by the use of AFFF. Despite the large range in documented BCF values, an average log BCF of 3 for edible parts can be used to quantify the transfer from surface water to fish (Table 1). Biomagnification factors have been established for animals higher up the trophic chain, like mink, eagle, walrus, etc. (Giesy & Kannan, 2001; Kannan et al., 2005; Tomy et al., 2004). However, since these animals are not part of the human food chain, these data are not included in this paper.

Pharmacokinetic models describing the accumulation of PFOS in various animal compartments (like liver, kidney and tissue) have been developed for rats and monkeys (Andersen, Clewell Iii, Tan,

Table 1
Overview of bioconcentration factors (BCFs) for various fish tissues.

Tissue	Country	Log BCF	Reference
Liver	Experimental study	3.7	Local contamination
	Japan	2.4–4.6	Background
	Canada	3.8–5.1	Local contamination
	USA	3.9	Background
Edible parts	USA	2.2–3.8	Background
	Experimental study	3.1	Boulanger, Vargo, Schnoor, and Hornbuckle (2004)
	Experimental study	2.5–2.9	3M (2003)
	Experimental study	3.0	OECD (2002) Martin et al. (2003)

Butenhoff, & Olsen, 2006; Tan, Clewell Iii, & Andersen, 2008). The same approach using other parameter values could be used to describe PFOS uptake by farm animals. For this purpose, data on PFOS levels in feed and farm animals are needed. Data for North Rhine Westphalia (the German pollution case) showed that only 4 out of 425 kidney samples from pigs showed PFOS levels >LOD with a maximum of 2.6 µg/kg. A higher percentage of kidney samples from cows (19 out of 248) contained positive PFOS levels up to 1332 µg/kg. A follow up study on cows (*n* = 18) showed that muscle tissue ranged from <LOD to 12.2 µg/kg, kidney samples from <LOD to 121 µg/kg and liver samples from 19.1 to 463 µg/kg (LANUV, 2008). Kidney and liver samples from cows outside the polluted area ranged from <LOD to 2.8 µg/kg (Brunner & Zimmermann, 2010). In a Japanese study, liver samples of farm animals ranged from 8.8 to 92 µg/kg. Serum levels were much lower ranging from 0.1 to 19 ng/ml (Guruge et al., 2008). However, since the exposure of these farm animals is unknown, transfer from the consumption of feed products and/or drinking water to the animals cannot be determined from these data. More research is, therefore, needed on the intake of PFOS by farm animals. One study reported on the oral uptake of PFOS in chicken (Yeung et al., 2009). PFCs were administered in a low (0.1 mg/kg bw) and high (1 mg/kg bw) dose for three weeks, after which blood, liver and kidney samples were taken. The remaining animals were allowed to depurate for a further 3 weeks after which the same samples were taken. The blood samples showed that PFOS concentrations increased from weeks 1 to 3 and accumulated at higher levels on week 4. The liver was the major reservoir for PFOS. However, at the end of the depuration phase the relative proportion of PFOS in the liver decreased whereas the proportion in the blood increased showing that PFOS is redistributed in the blood for removal from the body (Yeung et al., 2009). The data derived can be used to develop kinetic models on the oral uptake and elimination of PFOS in chicken blood and tissue. Data on transfer to edible tissues are not available, nor are comparable studies on other farm animals.

6. PFOS in food

Dietary intake is considered to be an important source of human exposure to PFOS (Fromme et al., 2009; Tittlemier et al., 2007; Trudel et al., 2008). However, information on the relative contributions of different food groups with respect to dietary intake is still quite limited (Halldorsson et al., 2008). Data are available for Sweden (Berger et al., 2007), Spain (Ericson, Martí-Cid, et al., 2008), Canada (Tittlemier et al., 2007), UK (Clarke et al., 2010; FSA, 2006), and USA (3M, 2001b) and are summarised in Table 2. This table shows that highest concentrations in positive samples are found in the food group of fruits and vegetables, which is mainly attributed to PFOS levels in potatoes up to 10 µg/kg (FSA, 2006). However, in a repetition of this study in the UK, PFOS in all potato samples remained below LOD (Clarke et al., 2010; FSA, 2009). The high values from the first study may have been caused by the sampling technique used. Inter laboratory studies revealed

Table 2
PFOS levels in various food groups.

Food group	<i>n</i>	%Pos	Mean level (µg/kg fresh weight) ^a			References ^b
			LOD = 0	LOD = 0.5LOD	>LOD	
Cereals	94	0.0%	0	3.97	–	1, 3–5
Dairy	113	5.3%	0.03	0.83	0.49	1–5
Fish	971	24.5%	0.66	1.41	2.68	1–10
Fruits and vegetables	324	22.8%	0.89	1.39	3.90	1, 3–5,7,8,11
Meat	399	16.8%	0.22	1.56	1.30	1–5,7,8
Poultry	176	29.6%	0.14	0.95	0.47	1–5,7,8
Other	98	0.0%	0	0.45	–	3,5,7,8

^a Mean levels calculated assuming that levels <LOD are either equal to 0 (first column) or equal to half the LOD (second column). The last column indicates mean levels calculated for positive samples.

^b Reference numbers refer to 1: (3M, 2001b), 2:(Berger et al., 2007), 3: (Clarke et al., 2010), 4: (Ericson, Martí-Cid, et al., 2008), 5: (FSA, 2006), 6: (So et al., 2006), 7: (Tittlemier et al., 2007), 8: (Jogsten et al., 2009), 9: (Nania et al., 2009), 10: (Gulowska et al., 2006), 11: (Knapp, 2008).

that sampling techniques and laboratory experience have increased since 2006 resulting in increased accuracy in monitoring results (Jahnke & Berger, 2009).

Table 2 shows that positive samples of both fish and meat contain high levels of PFOS averaging 2.7 and 1.3 µg/kg, respectively. Poultry had a higher percentage of positive samples than meat, but concentrations in the positive samples were lower. The levels of PFOS in fish may vary depending on the location. Data from the Sauerland region showed that fish from non-contaminated creeks contained less than 4 µg/kg PFOS, whereas fish from contaminated creeks contained levels up to 1180 µg/kg PFOS (Wilhelm et al., 2008). Dietary intake studies have indicated fish as an important source of dietary PFOS intake (Clarke et al., 2010; EFSA, 2008; Ericson, Martí-Cid, et al., 2008). However, a Canadian study showed that beef contributed around 90% of the total dietary intake, while fish was of lower importance (Tittlemier et al., 2007). This was confirmed in a Danish study showing that a high intake of meat relative to vegetables resulted in significant higher PFOS concentrations in human blood samples compared to a low intake of meat relative to vegetables. Based on this research, red meat was considered to be the most important source of dietary intake of PFOS. In contrast to other studies, the Danish study did not find a correlation with fish intake. The levels of PFOS in animal fats and snacks were also positively correlated to PFOS levels in human blood, where PFOS intake through snacks is probably related to leaching from food packaging (Halldorsson et al., 2008).

It is possible that in the general diet, red meat is the most important source, whereas in pollution events fish from contaminated waters may be the predominant source of human exposure (Wilhelm et al., 2008). As can be seen in Table 2 more data on levels of PFOS is available on fish than on meat samples. For a more accurate comparison between meat and fish contribution to PFOS intake, more data are needed on prevalence and concentrations present in the human diet (EFSA, 2008; Fromme et al., 2009).

7. Conclusions

Dietary intake has been shown to play an important role in human exposure to PFOS. In order to be able to effectively control and reduce the dietary intake of PFOS, it is essential to further investigate the fate of PFOS throughout the food chain. For this purpose, data are needed on PFOS prevalence and concentrations in the various steps from source to consumer together with possible transfer between the chain steps. This literature review shows that there are many data on levels of PFOS in surface water and fish. Since several papers have shown that levels in drinking water are comparable to surface water, these data can be used to determine PFOS intake through drinking water. Data of PFOS levels in soil, crops and animals are, however, scarce which hampers the quantification of transfer of PFOS from soil to animal products. Experiments focusing on uptake of PFOS from soil to crops, subsequent intake by animals and transfer into animal products will clarify the relevance of these steps in the food chain on human exposure. Field-based data on single pollution events can help to further elucidate the contribution of the various chain steps on the final level of PFOS in consumer products. In the well-documented cases of soil pollution in Sauerland and Alabama, analyses were indeed performed throughout the animal production chain starting in the polluted soil up to milk, feedstuffs and meat. The latter products showed low levels of PFOS, implying that intake from animal products would be of lesser importance (EPA, 2010; Wilhelm et al., 2008). However, it should be cautioned that, in these cases, the number of food samples taken was limited (e.g. only 2–4 milk samples were taken) and therefore, not sufficiently representative of PFOS transfer into food products. In case of environmental pollution events, it is thus advisable to not only monitor the polluted environment, but also include additional samples further along the food chain enabling the quantification of PFOS from farm-to-fork.

Once the transfer of PFOS through the food chain can be quantified, the relevance of each of the various chain steps can be assessed, facilitating research activities to focus on the most important chain steps. Furthermore, the effect of future pollution events on the consumer can be characterised using such parameterized food chain models, which allows the responsible authorities to take appropriate measures (if relevant) to protect consumer health.

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