



Environmental Property Modeling of Perfluorodecalin and its Implications for Environmental Fate and Hazards

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ABSTRACT

In a variety of medical and industrial uses, small amounts of perfluorodecalin ($C_{10}F_{18}$) may have been released into the environment. However, it may significantly contribute to the global warming due to its highly radiative efficiency and global warming potential (GWP). Using the chemical similarity approach, this article aimed at calculating the fate properties and discussing the environmental implications of the perfluorinated compound for the purpose of mitigating its emissions. The environmental fate properties of perfluorodecalin, including octanol-water partition coefficient, water solubility and Henry's law constant, were first estimated in the present study. These predicted values were further compared with those of other chemically similar compounds such as naphthalene, decalin and decane. From the computational findings, perfluorodecalin, which has exceptionally low solubility in water and high vaporization from the water bodies, tends to be hydrophobic and partitioned into organic matter, suggesting that it will sink into the atmosphere. Also addressed in the paper was a possible proposal for forming trifluoroacetic acid in the atmosphere by the ionized photolysis of perfluorodecalin.

Keywords: Perfluorinated compound; Atmospheric fate; Modeling; Environmental implication.

INTRODUCTION

Perfluorinated hydrocarbons or perfluorocarbons (PFCs), which contain only C-C and very strong C-F bonds (its binding energy 482.8 kJ/mol, compared with 410.0 kJ/mol for C-H bond and 323.0 kJ/mol for C-Cl bond), are recognized as specialty chemicals with unique physical and chemical properties (Kirsch, 2004). However, these synthetic fluorine-containing compounds are so effective absorbers of infrared radiation bands between 1000 and 1400 $1/cm$ that even small amounts significantly contribute to the global warming. Therefore, perfluorocarbons have been considered as one of the six target greenhouse gases under the Kyoto Protocol of the United Nations' Framework Convention on Climate Change (UNFCCC) in 1997. In the Protocol, the current target perfluorocarbons only aim at common perfluoro-n-alkanes, including gaseous and liquid compounds such as tetrafluoromethane (CF_4), hexafluoroethane (C_2F_6), octafluoropropane (C_3F_8), decafluoro-n-butane (C_4F_{10}), dodecafluoro-n-pentane (C_5F_{12}), and tetradecafluoro-n-hexane (C_6F_{14}) (IPCC, 2007).

Like liquid perfluoro-n-alkanes (C_nF_{2n+2} , $n = 5-9$) (Tsai, 2009), perfluorodecalin ($C_{10}F_{18}$) is a colorless, odorless, non-toxic, non-flammable, thermally stable, non-ozone-depleting, and heavy compound (high density and viscosity) with high volatility, low surface tension, high gas solubility, and very low solubility in water. Currently, it is primarily and increasingly used as a blood substitute (Lowe, 2008). In addition, it can be used as a contrast agent in a variety of diagnostic imaging techniques (e.g., ultrasound image) (Hall *et al.*, 2000), temporary intraoperative vitreous substitutes in vitreoretinal surgery (ophthalmology) (Heimann *et al.*, 2008), cosmetic ointment additive for repairing burned skin and wound surface (Oxynoid *et al.*, 1994), liquid ventilation used in the drug delivery (Kraft, 2001), carrier of glassified microspheres that contain vaccines (Coghlan, 2004), reaction medium in organic and organometallic syntheses (Hibbert *et al.*, 1997; Sandford, 2003), volatile surfactant used for gas modification of lubricants and in optics and liquid lasers (Stoilov, 1998), and tracer gas in the environmental quality modeling in the ocean, and groundwater (Watson *et al.*, 1987; Deeds *et al.*, 1999).

Before 2005, the global warming potential (GWP) of perfluorodecalin was not available, making it difficult for medical users and regulating bodies to assess the environmental risks. It was even considered not to be a potent greenhouse gas although perfluorodecalin is expected

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to have an atmospheric lifetime on the order of 1000 years. However, the updated data on the radiative efficiency and GWP (100 year time horizon) for perfluorodecalin assuming a lifetime of 1000 years indicate 0.56 W/m²/ppbv and 7200 times more potent than carbon dioxide (CO₂) in trapping solar heat, respectively (Shine *et al.*, 2005). Also pointed out by Watson *et al.* (2007), the trace concentrations of perfluorodecalin and other perfluorocarbon compounds in the atmosphere trivially contribute to climate change and other environmental effects at current release rates (of order 10 tonnes per year) (Simmonds *et al.*, 2002; Shine *et al.*, 2005), but it would have more significant impacts on the global warming in the future.

From the viewpoint of molecular structures, perfluorodecalin (C₁₀F₁₈) is a derivative of decalin (C₁₀H₁₈) in which all of the hydrogen atoms are replaced by fluorine atoms. In order to strengthen our understanding of such potent IR-absorbing compound for mitigating its emissions, it is vital to investigate the environmental fate and transport between interfaces like air, water, and solid phases for C₁₀F₁₈. However, the measured properties regarding the chemical distribution among phases are very scarce in the literature (Dias *et al.*, 2005). This paper aimed to present the preliminary predications of octanol-water partition coefficient, water solubility, and Henry's law constant for C₁₀F₁₈. Furthermore, the comparison between the environmental fate and implications of perfluorodecalin (C₁₀F₁₈) and other perfluorinated compounds was discussed.

METHODS

In the discussion of environmental distribution among phases for a given compound, it is hypothesized that the target compound is not chemically transformed. In this respect, perfluorodecalin has been considered to be extremely stable, highly inert and unlikely decomposed under the normal environmental conditions. It means that the environmental fate of the fully fluorinated molecule if released or emitted is only transported from a phase to another without changing its identities until the equilibrium approached. Therefore, the properties regarding the environmental distribution among air, water, and solid phases are considerably important in understanding its movement between media and evaluating its behavior within a single medium. These distribution properties commonly include octanol-water partition coefficient (K_{ow}), water solubility (S), Henry's law constant (K_H), and vapor pressure (P). Recent studies on its environmental properties were focused on its vapor pressure (P , kPa) with temperature (T , K) (Dias *et al.*, 2005), and given as follows:

$$\ln P = 15.938 - \frac{4525.811}{T - 15.354} \quad (1)$$

Therefore, the estimation of K_{ow} , S and K_H for C₁₀F₁₈ based on the chemical's structure was described below. By contrast, the measured or observed data on K_{ow} , S and K_H of decalin (C₁₀H₁₈) and other chemically similar

compounds at 298 K in the literature were also discussed in the work.

In the present paper, the value of partition coefficient for perfluorodecalin was preliminarily estimated using the "fragment constant" approach in which the target compound was divided into fragments and values of each group are estimated together (Meylan and Howard, 1995),

$$\log K_{ow} = 0.229 + \sum (m_i \times f_i) \quad (2)$$

where $\log K_{ow}$ is the base 10 logarithm of the ratio the chemical's concentration in octanol to the chemical's concentration in water, m_i is the number of groups of type i in the molecule, f_i is the contribution of each group to the partition coefficient, and the summation is taken over all groups. Referring to the contribution groups in the method, perfluorodecalin can be represented by ten > C < groups and eighteen – F (aliphatic attachment) groups. Because the octanol/water partition coefficient (K_{ow}) is related to the solubility of a chemical in water (denoted as S ; unit: $\mu\text{mol/L}$), there are different regression equations (i.e., $\log S = a \log K_{ow} + b$) for the estimation of water solubility in the literature (Lyman *et al.*, 1990). It showed that a plot of $\log S$ vs. $\log K_{ow}$ has a slope of about -1 (i.e., $a \doteq -1$). For example, a slope of -0.962 has been correlated with halogenated hydrocarbons.

On the other hand, the Henry's law is used to describe the partition of a gas in two different phases, such as water and air, under equilibrium conditions. Therefore, the Henry's law constant (K_H) is a partition coefficient, which is usually defined as the ratio of a chemical's concentration in air to its concentration in water at equilibrium, or mathematically described as

$$\lim \left(\frac{f_2}{x_2} \right)_{x_2 \rightarrow 0} = K_H \quad (3)$$

where x_2 is the mole fraction of the solute gas in aqueous solution and f_2 is the fugacity of the solute gas. For dilute solution at low pressure, the above equation is then written as

$$K_H = \frac{P_2}{x_2} \quad (4)$$

where P_2 is the partial pressure of the solute gas over the solution. In the paper, P_2 is expressed in atmospheres (Pa), and x_2 as a mole fraction. Thus, K_H has units of Pa. For very stable compounds, K_H can be proximately estimated by dividing the vapor pressure of a target gas by its water solubility at the same temperature (e.g., 298 K) if it has not yet been measured or observed in the literature (Lyman *et al.*, 1990).

RESULTS AND DISCUSSION

Partition Coefficient

It is well known that several methods are available for the estimation of K_{ow} of organic chemicals (Lyman *et al.*, 1990).

However, these approaches are somewhat complicated. The target compound, C₁₀F₁₈, is a newly synthesized compound only containing carbon and fluorine. Thus, there were no measured data on its distribution properties in the literature. In the present work, the values of coefficient f_i for C₁₀F₁₈ were obtained from the literature (Meylan and Howard, 1995). The estimated values of $\log K_{ow}$ for C₁₀F₁₈ can be thus calculated by the following equation

$$\log K_{ow} = 0.229 + 10 \times 0.2676 + 18 \times (-0.0031) \doteq 2.85$$

The predicted value of $\log K_{ow}$ for C₁₀F₁₈ is below 3.5, showing that it has medium potential bioaccumulation in the environment (Allen and Shonnard, 2002). To understand the availability of the method in the estimation of partition coefficients for C₁₀F₁₈, three hydrocarbons which have the same carbon number (i.e., naphthalene C₁₀H₈, decalin C₁₀H₁₈, and decane C₁₀H₂₂) and are similar to C₁₀F₁₈ in chemical structure were estimated to obtain their partition coefficients, which were then compared to those reported or measured in the literature (Yaws, 2008). According to their molecular structures, the predicted values of $\log K_{ow}$ are given by:

$$\begin{array}{ll} \text{naphthalene} & \log K_{ow} = 0.229 + 10 \times 0.2940 \doteq 3.17 \\ \text{decalin} & \log K_{ow} = 0.229 + 2 \times 0.3614 + 8 \times 0.4911 \\ & \doteq 4.88 \\ \text{decane} & \log K_{ow} = 0.229 + 2 \times 0.5473 + 8 \times 0.4911 \\ & \doteq 5.25 \end{array}$$

The experimental or reported values of naphthalene, decalin and decane are 3.34, 4.20 and 6.25, respectively. Therefore, the predicted values of $\log K_{ow}$ for the three hydrocarbons are in errors within 20%, indicating that the Meylan & Howard method may be practically useful for predicating the octanol-water partition coefficient for perfluorodecalin. Of course, it would be helpful in measuring K_{ow} in the laboratory to compare the estimated value of partition coefficient for C₁₀F₁₈.

Water Solubility

Of the various fate and transport properties that affect the environmental distribution between air and water, water solubility is one of the most important parameters. By using the measured data on the water solubility of CF₄ in 298 K (i.e., 2.1×10^{-4} mol/L, or 3.8×10^{-6} in mole fraction) (Cosgrove and Walkley, 1981) and the value of $\log K_{ow}$ (i.e., 1.18) (Yaws, 2008), the water solubility (S) of perfluorodecalin at 298 K can be roughly estimated as follows (Lyman *et al.*, 1990):

$$\begin{aligned} \log (S/2.1 \times 10^{-4}) &\doteq -(2.85 - 1.18) \\ S &\doteq 4.5 \times 10^{-6} \text{ mol/L or } 8.0 \times 10^{-8} \text{ in mole fraction} \end{aligned}$$

This prediction led to the conclusion that perfluorodecalin has lower solubility in water than the fully fluorinated gases such as CF₄ and SF₆ (water solubility at 298 K is equal to 2.47×10^{-4} mol/L, or 4.44×10^{-6} in mole fraction) (Cosgrove and Walkley, 1981). The result is consistent

with its molecular structure because of the extremely nonpolar character and very low attraction forces between the perfluorodecalin molecules.

Henry's Law Constant

The predicted value of $\ln (K_H/\text{MPa})$ for C₁₀F₁₈ was thus estimated to be about 9.3 based on its vapor pressure (i.e., 0.91 kPa, at 298 K) (Dias *et al.*, 2005) and the predicted solubility (i.e., 8.0×10^{-8} in mole fraction) in water described above. The predicted value implied that C₁₀F₁₈ has high vaporization from water bodies to atmosphere. On the other hand, the predicted value of K_H for C₁₀F₁₈ is significantly lower than the measured value (i.e., $\ln (K_H/\text{MPa}) = 10.04$) of K_H for SF₆ at 298 K (Mroczek, 1997). It was also noted that C₁₀F₁₈ is very stable in the atmosphere, implying that it does not react with highly oxidative species (e.g., O₃) and free radicals such as hydroxyl radical and it can not be decomposed by direct or sensitized photolysis, either. Therefore, this potent greenhouse gas will accumulate in the troposphere and even in the stratosphere.

Environmental Implications of Perfluorodecalin

Fluorinated alkanes are inert to most chemical attacks because their chemical reactivities decrease with increasing substitution of fluorine for an organic molecule until fluorination is completed. Consequently, fully fluorinated compound has an atmospheric lifetime of a few thousand years or more (Ravishankara *et al.*, 1993). Based on the updated data in Table 1 (IPCC, 2007), it is clear that liquid perfluoro-n-alkanes are potent greenhouse gases as compared to other gaseous perfluoro-n-alkanes such as CF₄ and C₂F₆. Because of its extremely chemical stability, perfluorodecalin is expected not to be degraded in the tropospheric environment by reactions with highly oxidative species (e.g., O₃) and free radicals such as hydroxyl radical and O(¹D) (Ravishankara *et al.*, 1993). It was thus suggested that photolysis at high-energy radiation (e.g., 121.6 nm) in the upper layers, or ion-molecule reactions occurring in the electrical spark can have an effect on the removal of C₁₀F₁₈ from the atmosphere (Morris *et al.*, 1995).

As mentioned above, perfluorodecalin does not contain chlorine and is hence not ozone-depleting substance. Also, it hardly contributes to the formation of photochemical oxidants in the troposphere, and it is not removed by wet deposition via rainfall and dry deposition to the surface due to its predicted fate properties - extremely low water solubility and high fugacity from water bodies to atmosphere. With respect to its gas-phase photochemistry of C₁₀F₁₈ in the atmosphere, a proposal for a possible and reasonable sequence of steps connecting the formation of CF₃COOH was addressed in the present paper. For example, the ionization of C₁₀F₁₈ may be initiated by reaction with reactive ions (e.g., O⁺, O₂⁺, O₂⁻, and H₃O⁺) under thermal conditions such as pyrolysis and lightning. This is similar to the thermal decomposition of perfluorocompound (e.g., BF₃) in the plasma environment (Wang *et al.*, 2003). The resulting fully fluorinated radical, CF₃CF₂·, could further react with hydroxyl radical (OH·) to form more stable

Table 1. Some atmospheric status and global warming of perfluorodecalin and other perfluorocompounds.^a

Compound	Atmospheric concentration (ppt)	Radiative efficiency (W/m ² /ppb)	Atmospheric lifetime (years)	GWP (100 yr time horizon)
SF ₆	4.2	0.52	3,200	22,450
CF ₄	74	0.10	50,000	7,390
C ₂ F ₆	2.9	0.26	10,000	12,200
C ₅ F ₁₂	<1.0 ^b	0.41	4,100	9,160
C ₆ F ₁₄	<1.0 ^b	0.49	3,200	9,300
C ₁₀ F ₁₈	<0.10 ^c	0.56	>1,000	>7,500

^a Most data were compiled from the reference (IPCC, 2007); ^b Estimated by the author; ^c Reported by Simmonds *et al.* (2002).

molecule, CF₃CF₂OH (Wallington *et al.*, 1994). The remaining steps in the photochemical reactions involve the elimination of HF and the formation of CF₃COF. It has been shown that CF₃COF is preferably dominated by incorporation into cloud water followed by rapid hydrolysis to trifluoroacetic acid (CF₃COOH, TFA) and the elimination of HF (Ellis *et al.*, 2004). From the proposed outcome, it was very similar to atmospheric oxidation of HFC-134a (1,1,1,2-tetrafluoroethane), which can be decomposed to CF₃COF and thence TFA, or trifluoroacetate ion in the aqueous environment (Wallington *et al.*, 1994). TFA is a strong organic acid with high miscibility in water. Its low octanol/water partition coefficient (log K_{ow} = -2.1) indicates no potential to bioaccumulation (Boutonnet *et al.*, 1999). It should also be noted that hydrogen fluoride or dissociated fluoride ions may be formed when medical devices containing perfluorocarbons are sterilized by gamma and beta irradiation (Zundorf *et al.*, 2008). On the other hand, however, that the decomposition products of perfluorodecalin, pyrolyzed by electrical and thermal decomposition under thermal conditions, are probably toxic and even corrosive. The pyrolysis products include tetrafluoroethylene (C₂F₄), hexafluoropropylene (C₃F₆) and perfluoroisobutylene (C₄F₈) (O'Mahony *et al.*, 1993). This situation is very similar to those where some toxic products can be evolved from polytetrafluoroethylene (Teflon) pyrolysis processes at high temperature (Tsai *et al.*, 2000). Among these toxic decomposition products, perfluorobutylene, CF₂ = C(CF₃)₂, is a highly toxic gas, which can cause acute lung injury (pulmonary edema) due to its rapid hydrolysis in contact with water, producing various reactive compounds and fluorophosgene (COF₂). Another decomposition product, tetrafluoroethylene (CF₂ = CF₂), does not pose acute toxic hazards, but has a chronic toxicity. It should be noted that the carcinogenicity of tetrafluoroethylene has been recognized as "A3-confirmed animal carcinogen with unknown relevance to humans" by the American Conference of Industrial Hygienists (ACGIH) and as "Group B2- possibly carcinogenic to humans" by the International Agency for Research on Cancer (IARC) (IARC, 2011).

CONCLUSIONS

Although perfluorodecalin (C₁₀F₁₈) addressed in this paper seemed to be included in the basket of the Kyoto Protocol, its emissions from various medical/industrial

applications were rarely described in the National Inventory Reports by Annex I Parties. Noticeably, the liquid perfluorocarbons still pose some hazards to the environment, especially to global warming due to its atmospheric lifetime. In the paper, the analysis of the environmental fate of perfluorodecalin and its atmospheric implications indicated that it does not sink into the environment at normal conditions according to the predicted values of partition coefficient, water solubility and Henry's law constant. Also addressed in the paper was a possible proposal for forming trifluoroacetic acid (CF₃COOH) in the atmosphere by the ionized photolysis of perfluorodecalin. However, its impacts on the global climate change may be more significant in the future because of its long lifetime in the atmosphere and its various medical uses.

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