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Fuzzy synthetic evaluation of disinfection by-products – a risk-based indexing system

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Abstract: Disinfection by-products (DBPs) are formed when disinfectants such as chlorine, chloramine, and ozone react with organic matter in water. Chlorine being the most common disinfectant used in the drinking water industry worldwide, significant attention has been focused on chlorinated DBPs. An new indexing method using fuzzy synthetic evaluation is proposed to determine the health risk associated with the two major groups of chlorinated DBPs - trihalomethanes (THMs) and haloacetic acids (HAAs). Initially, membership functions for cancer and non-cancer risks associated with THMs and HAAs are used to establish the fuzzy evaluation matrices. Subsequently, weighted evaluation matrices for both types of risks are established by performing cross products on the weighted vectors (founded on analytic hierarchy process) and the fuzzy evaluation matrices. In the final stage, the weighted evaluation matrices of cancer and non-cancer risks are aggregated to determine the final risk rating. Two case studies are provided to demonstrate the application of this method.

Keywords: Water quality, DBPs, fuzzy synthetic evaluation, AHP, cancer and non-cancer risk.

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1. INTRODUCTION

The disinfection is performed to eradicate and/or inactivate the pathogens from drinking water. The disinfection involves destruction of organization of cell structure, and interference with energy yielding metabolisms, biosynthesis and growth of microbes. Chlorine and its compounds are the most commonly used disinfectants for water treatment. Chlorine has a very strong oxidizing potential, which provides a residual throughout the distribution system and protects against microbial recontamination.

The addition of disinfecting chemicals to drinking water can reduce the microbial risk but poses chemical risk due to the formation of disinfection by-products (DBPs). Formation of DBPs occurs when the disinfectant reacts with natural organic matter and/or inorganic substances present in water. More than 600 DBPs have been identified in the laboratory scale studies for different disinfectants, while approximately 250 have been identified in drinking water samples taken from actual distribution systems. Among DBPs found in chlorinated drinking water, trihalomethanes (THMs) and haloacetic acids (HAAs) have been the focus of particular attention because of the potential carcinogenicity and harmful non-cancer effects. THMs include four compounds – chloroform (TCM), dichlorobromomethane (DCBM), dibromochloromethane (DBCM) and bromoform (TBM). In waters of low bromide levels, TCM is a major culprit among THMs. HAAs include nine compounds but the most common are – monochloroacetic acid (MCAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), monobromoacetic acid (MBAA), dibromoacetic acid (DBAA), and bromochloroacetic acid (BCAA) (Serodes *et al*., 2003). Among them, generally DCAA and TCAA are of significant levels in chlorinated drinking water.

The major objective of this research is to develop a risk-based indexing system for chlorinated DBPs found in drinking water using *fuzzy synthetic evaluation* (FSE) technique. The paper addresses three major issues: identifying toxicity criteria for of DBPs, developing an indexing system by grouping DBPs based on associated adverse health effects, and demonstrating the application of this method using two case studies.

1.1. TOXICITY OF THMS AND HAAS

The Safe Drinking Water Act requires the United States Environmental Protection Agency (US EPA) to develop new drinking water regulations. The regulations related to DBPs are part of the Microbial-Disinfection by-products (M-DBPs) rule (US EPA, 1999). The DBP regulations are based on evidence of their potential adverse human health effects, in particular cancer and reproductive disorders (Cantor *et al*., 1998; Graves *et al*., 2002). Routine water quality sampling help in identifying whether regulatory thresholds (guideline or standard) of DBPs are violated or not. The threshold values are based on potential toxicity of DBP indicators. A wealth of literature reporting adverse health effects through toxicological laboratory studies is available. Some of the adverse health effects of THMs and few HAAs are summarised in Table 1.

The World health Organization (WHO, 1993) published drinking water guidelines for a few common DBPs including THMs and HAAs. In addition to guidelines for THMs, the WHO has also suggested that *the sum of the ratios of the THM levels to the guideline values should not exceed 1* (see Table 2). Such guidelines have no official recognition in the US or Canada. The US EPA (2001) has established the maximum allowable contaminant level of 0.08 mg/L for total THMs and of 0.06 mg/L for HAA_5 (the sum of five HAAs, that is mono-, di-, and trichloroacetic acids and mono- and dibromoacetic acids), respectively. Compliance of these by-products is based on an annual running average of quarterly samples, and since 2002 will also be based on a locational running average (Sharfenaker, 2001). Health Canada (2001) has set total THM levels of 0.10 mg/L as an interim maximum acceptable concentration, which serves as a guideline for Provincial regulations. No Canadian drinking water quality guideline exists for other DBPs for the time being. The Australian-New-Zealand (2000) and UK (2000) drinking water standards are also summarized in Table 2 for comparison.

1.2. WATER QUALITY INDEXING

Water quality is generally defined by upper and lower limits on selected possible contaminants in water (Maier, 1999). Traditionally, water quality indicators (or parameters) can be grouped into three broad categories - physical, chemical and biological, and each category contains a number of water quality variables. The acceptability of water quality for its intended

use depends on the magnitude of these indicators (Swamee and Tyagi, 2000), and is often governed by regulations. A water quality failure is often defined as *an exceedence of one or more water quality indicators (DBPs) from specific regulations, or in the absence of regulations, exceedence of guidelines or self-imposed, customer-driven limits*.

Recently, significant literature is published on describing the overall (aggregate) water quality by an index using various statistical and mathematical techniques. Swamee and Tyagi (2000) have discussed in detail the pros and cons of different techniques and approaches available for evaluating the water quality index. Sinha *et al*. (1994) used pH, chloride concentration, turbidity, residual chlorine, conductivity and MPN (Most probable number – a bacterial counting technique) into a single water quality index through a weighting scheme, which can represent an overall water quality at various nodes in the distribution system. The normalized water quality index (0-100) defines the overall water quality in each segment of the distribution system.

Sadiq *et al*. (2003, 2004a) have recently suggested a fuzzy-based framework for the analysis of aggregative risk associated with water quality failure in the distribution system. The basic risk items are grouped into higher level risk factors, to form a multi-stage hierarchical model of aggregative risk for water quality failure in the distribution network. The usage of fuzzy set techniques for water quality indexing enables the incorporation of hard field data (e.g. observed water quality) and soft qualitative data (e.g. expert opinion).

1.3. FUZZY SETS AND SOFT COMPUTING

The term soft computing describes an array of emerging techniques such as fuzzy logic, probabilistic reasoning, neural networks, and genetic algorithms. All these techniques are essentially heuristic which provide rational and reasoned out solutions for complex real-world problems (Bonissone, 1997). Quantitative aggregation of risk due to multiple sources is a complex process, which warrants soft computing techniques.

Fuzzy logic provides a language with syntax and semantics to translate qualitative knowledge into numerical reasoning. In many engineering problems, the information about the probabilities of various risk items is vaguely known or assessed. The term *computing with words*

has been introduced by Zadeh (1996) to explain the notion of reasoning linguistically rather than with numerical quantities. Such reasoning has a central importance for many emerging technologies related to engineering and applied sciences. This approach has proved very useful in medical diagnosis (Lascio *et al*., 2002), information technology (Lee, 1996), water quality assessment (Lu *et al*., 1999; Lu and Lo, 2002), corrosion of cast iron pipes (Sadiq *et al*., 2004b) and in many other industrial applications (Lawry, 2001).

When evaluating risk items in complex systems, decision-makers, engineers, managers, regulators and other stake-holders often view risk in terms of linguistic variables like *very high*, *high*, *very low*, *low* etc. The fuzzy set theory is able to deal effectively with uncertain, vague and linguistic variables, which can be used for approximate reasoning and subsequently manipulated to propagate the uncertainties throughout the decision process. Fuzzy-based techniques are a generalized form of interval analysis used to address uncertain and/or imprecise information. A fuzzy set describes the relationship between an uncertain quantity x and a membership function μ, which ranges between 0 and 1. A fuzzy set is an extension of the traditional set theory (in which x is either a member of set A or not) in that an x can be a member of set A with a certain degree of membership μ. To qualify as a fuzzy number, a fuzzy set must be normal, convex and bounded (see Klir and Yuan (1995) for definitions of these terminologies). Any shape of a fuzzy number is possible, but the selected shape should be justified by available information. Generally, triangular fuzzy numbers (TFN) or trapezoidal fuzzy numbers (ZFN) are used for representing linguistic variables (Lee, 1996). *Defuzzification* is a process to evaluate a crisp or point estimate of a fuzzy number. A defuzzified value is generally represented by the centroid, often determined using the centre of area method (Yager, 1980).

Fuzzy-based techniques can help in addressing deficiencies inherent in binary logic and are useful in propagating uncertainties through models. Contrary to binary logic, fuzzy-based techniques can provide an *intensity* of exceeding regulated thresholds with the help of memberships to various risk levels. In water quality modeling, the fuzzy set theory has been used for classification of rivers since 1980s. The majority of research has been focused on fuzzy synthetic evaluation (FSE) and fuzzy clustering analysis (FCA). The FSE is used to classify samples at a known centre of classification (or group), whereas the FCA is used to classify samples according to their relationships when this centre is unknown (Lu *et al*., 1999). The FSE

classifies samples for known standards and guidelines, which is a modified version of traditional synthetic evaluation techniques.

2. FUZZY SYNTHETIC EVALUATION FOR DBPS

The US EPA (2003) has classified various chemicals based on their carcinogenicity potential and other detrimental effects. Generally the environmental databases report either the slope factors (*SF*) or concentrations corresponding to unit risk for different routes of exposures for carcinogenic compounds. Toxicity data are obtained through laboratory experiments and epidemiological studies. Either extrapolation models or uncertainty (and modifying) factors are used to convert animal toxicity data like no-observed- allowable-effect-concentration (*NOAEL*) and low-observed-allowable-effect-concentration (*LOAEL*) to estimate reference dose (*RfD*) for non-cancer risk effects. The threshold concentration for non-cancer risk effects can be determined from risk quotient ($RQ = 1$), which is the ratio of exposure concentration (converted into dose based on exposure time) to *RfD*. In this paper, four species of THMs and two HAAs are selected for the fuzzy synthetic evaluation of drinking water in distribution system (see Table 3).

The three-step process, of *fuzzification*, *aggregation* and *defuzzification* is commonly used for fuzzy-based decision-making (Lu *et al*., 1999). This approach is employed here to translate observations to risk levels, and is referred to as *fuzzy synthetic evaluation* (FSE).

2.1. MEMBERSHIP FUNCTIONS FOR CANCER AND NON-CANCER RISKS - FUZZIFICATION

DBPs have potential to cause human cancer and non-cancer risks through consumption of drinking water. The four species of THMs - TCM, DBCM, BDCM and TBM - and two haloacetic acids – DCAA and TCAA – are used in this analysis. For both cancer and non-cancer risks the triangular fuzzy numbers (TFNs) are defined using three partitions – *low*, *medium* and *high risk* as shown in Table 4. For cancer effects, the *low risk* is defined as *less than one in a million* (10^{-6}) and the *high risk* is defined as *more than one in ten thousands* (10^{-4}). The intermediate range is defined as *medium risk*. To fuzzify this information the TFN for *low risk* is defined such that the membership μ of 0.0 is assigned at the midpoint of the *medium risk*. Similarly, for *high risk* TFN the μ of 0.0 is assigned at the midpoint of the *medium risk*. The membership of *medium risk* over the universe of discourse is defined as

$$\mu_M = 1 - \mu_L - \mu_H \quad (1)$$

For non-cancer effects, the *low risk* is defined for concentration at risk quotient $RQ \leq 0.01$ and *high risk* is defined for concentration of $RQ > 1$. The intermediate range is defined as *medium risk*. To fuzzify this information, the TFN for *low risk* is defined such that the membership μ of 0.0 is assigned at the midpoint of the *medium risk* (concentration corresponding to *RQ* of 0.1). Similarly, for *high risk* TFN, the μ of 0.0 is assigned at the midpoint of the *medium risk*. The membership for *medium risk* over the universe of discourse is estimated using equation 1 (see Table 4).

2.2. WEIGHTING SCHEME

Fuzzy synthetic evaluation requires information for relative importance of attributes or criteria (e.g., for various DBPs species). The relative importance is established by a set of preference weights, which can be normalized to a sum of 1. In case of *n* criteria, a set of weights can be written as

$$W = (w_1, w_2, ..., w_n) \quad \text{where} \quad \sum_{j=1}^{n} w_j = 1 \quad (2)$$

Saaty (1988) proposed an analytical hierarchy process (AHP) to estimate the relative importance of each attribute (in a group) using pair-wise comparisons. Lu *et al*. (1999), Sadiq *et al*. (2004b), and Khan *et al*. (2002) also used a similar technique for calculating the weights of multiple attributes. The relative importance of different factors is assigned using intensity of importance using factors from 1 to 9, where 1 represents "*equal importance*" and 9 represents "*extreme importance*" (see Saaty (1988) for detail). An importance matrix, *J*, can be established, where each element, j_{mn}, in the upper triangular matrix expresses the importance intensity of a criterion (or property) *m* with respect to another criterion *n*. For example, in the importance matrix, *J*, below, chloroform has been assigned importance intensities 2 and 3 times greater than DBCM and BDCM, respectively for cancer effects. Each element in the lower triangle of the matrix is just the reciprocal of an element in the upper triangle, i.e., $j_{nm} = 1/j_{mn}$. The importance matrix *J*, for cancer risk was thus developed as an example:

$$J = \begin{array}{c} \\ \text{TCM} \\ \text{DBCM} \\ \text{BDCM} \\ \text{TBM} \\ \text{DCAA} \\ \text{TCAA} \end{array} \begin{array}{c} \begin{array}{cccccc} \text{TCM} & \text{DBCM} & \text{BDCM} & \text{TBM} & \text{DCAA} & \text{TCAA} \end{array} \\ \begin{bmatrix} 1 & 2 & 3 & 1 & 1 & 2 \\ 0.5 & 1 & 1.5 & 0.5 & 0.5 & 1 \\ 0.33 & 0.67 & 1 & 0.33 & 0.33 & 0.67 \\ 1 & 2 & 3 & 1 & 1 & 2 \\ 1 & 2 & 3 & 1 & 1 & 2 \\ 0.5 & 1 & 1.5 & 0.5 & 0.5 & 1 \end{bmatrix} \end{array} \tag{3}$$

The importance value of each element, j_{mn}, in J matrix above should be assigned based on expert opinion on how the different DBPs species affect human health under the specific circumstances. In this case, the US EPA (2003) carcinogenicity potential ranking system is used to assign importance factors (see Table 3). Members j_{mn} can be modified as required if better information becomes available. A matrix I can be determined by taking the geometric mean of each row and then the weighted vector W can be derived by normalization of matrix I.

$$I = \begin{bmatrix} 1.51 \\ 0.76 \\ 0.50 \\ 1.51 \\ 1.51 \\ 0.76 \end{bmatrix} \Rightarrow W_{cancer} = \begin{bmatrix} w_{TCM} \\ w_{DBCM} \\ w_{BDCM} \\ w_{TBM} \\ w_{DCAA} \\ w_{TCAA} \end{bmatrix} = \begin{bmatrix} 0.231 \\ 0.115 \\ 0.077 \\ 0.231 \\ 0.231 \\ 0.115 \end{bmatrix} \tag{4}$$

The weighted vector W_{cancer} indicates that TCM, TBM and DCAA will contribute more to overall cancer risk because of their higher carcinogenicity potential (defined as B2 in US EPA ranking system). Similarly, the importance matrix (J) is established for non-cancer risk and the weights for non-cancer risk are estimated. The weights estimated (for DBPs) causing cancer and non-cancer risks are summarized in Table 5. In the second stage, the final weights for cancer and non-cancer risks are estimated by assigning cancer risk 2 times more importance than the non-cancer risks (Lee, 1992). The aggregation process is shown in Figure 1.

2.3. AGGREGATION

Table 4 is used to establish the membership for TFNs of cancer and non-cancer risks caused by consumption of drinking water. As described before, risk in each case is defined by three partitions - *low* (L), *medium* (M), and *high* (H). The memberships of these partitions (risk levels) are used to establish an evaluation matrix *A,* where each row represents levels of cancer (or non-cancer) risk due to various DBPs. The weight vector $w_{cancer} = [w_{TCM} \quad w_{DBCM} \quad w_{BDCM} \quad w_{TBM} \quad w_{DCAA} \quad w_{TCAA}]$ is multiplied (cross product) by matrix *A* to determine matrix B_{cancer}.

$$\left.\begin{aligned} B_{cancer} &= w_{cancer} \times A_{cancer} = \begin{bmatrix} b_C^L & b_C^M & b_C^H \end{bmatrix} \\ B_{cancer} &= \begin{bmatrix} w_{TCM} & w_{DBCM} & w_{BDCM} & w_{TBM} & w_{DCAA} & w_{TCAA} \end{bmatrix}_{Cancer} \times \begin{bmatrix} \mu_{TCM}^L & \mu_{TCM}^M & \mu_{TCM}^H \\ \mu_{DBCM}^L & \mu_{DBCM}^M & \mu_{DBCM}^H \\ \mu_{BDCM}^L & \mu_{BDCM}^M & \mu_{BDCM}^H \\ \mu_{TBM}^L & \mu_{TBM}^M & \mu_{TBM}^H \\ \mu_{DCAA}^L & \mu_{DCAA}^M & \mu_{DCAA}^H \\ \mu_{TCAA}^L & \mu_{TCAA}^M & \mu_{TCAA}^H \end{bmatrix} \end{aligned}\right\} \quad (5)$$

Similarly for non-cancer risk the matrix $B_{non\text{-}cancer}$ will be

$$B_{non-cancer} = w_{non-cancer} \times A_{non-cancer} = \begin{bmatrix} b_{NC}^L & b_{NC}^M & b_{NC}^H \end{bmatrix}$$

The final fuzzy evaluation matrix for risk *R* can be determined as

$$R = \begin{bmatrix} w_{Cancer} & w_{Non-cancer} \end{bmatrix} \times \begin{bmatrix} b_C^L & b_C^M & b_C^H \\ b_{NC}^L & b_{NC}^M & b_{NC}^H \end{bmatrix} = \begin{bmatrix} r^L & r^M & r^H \end{bmatrix} \quad (6)$$

2.4. DEFUZZIFICATION

The contribution of each DBP in the aggregation process provides only a "partial evidence" to the intensity of final risk. Though the memberships of DBPs to *low*, *medium* and *high* risks are monotonic over the universe of discourse, but it does not guarantee that the final fuzzy set for risk will maintain that sequence, especially in the extreme cases where some parameters have "high value" memberships to *low* risk, and others have "high value" memberships to *high* risk. To make this process more meaningful and intuitive, *defuzzification* is

performed. It can be achieved through *maximising* risk membership in matrix R (Cheng and Lin, 2002).

$$K = max\left(r^L, r^M, r^H\right) \tag{7}$$

K represents the predominant risk level i.e. the highest value of membership, which decides the overall risk classification. The crisp value of risk can also be determined by assigning weights to memberships of the risk matrix (Lu *et al*., 1999; Silvert 2000). For example, the following equation is used for defuzzification in this study:

$$Risk\ index = RI = 0.5 \times r^L + 1 \times r^M + 2.3 \times r^H \tag{8}$$

The coefficients (weights) are assigned arbitrarily in this study and guidelines may be established for risk-index (Lu *et al*., 1999). To make the risk-index more meaningful, the coefficients are adjusted so that the risk-index matches the threshold suggested by regulatory agencies. The US EPA (2001) has suggested maximum contaminant levels for total THMs (80 µg/L) and HAAs (60 µg/L). The water will be declared unfit for drinking if any of the ratios will exceed 1.0. The water will be considered safe for drinking if the following equation holds:

$$Std.\ Method = max\left[\left(\frac{\sum Concentration\ of\ THMs}{80}\right), \left(\frac{\sum Concentration\ of\ HAAs}{60}\right)\right] \leq 1.0 \tag{9}$$

For example if the standard method (equation 9) ratio is estimated at 0.5, it implies that the concentration of either THMs or HAAs (which ever is the highest) is half of the threshold (guideline value). The *maximum* operator is used because of its conservativeness. Though if the ratio exceeds 1 (regardless of how big is the ratio), implication is that the regulatory thresholds are violated.

To make proposed index more meaningful, the risk-index estimated by fuzzy synthetic evaluation technique can be linked to standard method ratio using simple regression analysis:

$$Std.\ Method = f(RI) \tag{10}$$

3. APPLICATION

The fuzzy synthetic evaluation procedure described in section 3 was applied to two case studies in the Quebec City region (Canada). The application relies on data on chlorinated DBPs generated under experimental chlorination of waters of two utilities: Quebec City and Sainte-Foy. The data represent the potential occurrence of DBPs in the distribution systems of the utilities. The Quebec City utility takes the water from a river with high colour content while Sainte-Foy utility takes the water from the Saint-Lawrence River, which is low in colour but contains relatively high turbidity. The water treatment process is similar for both utilities (pre-oxidation, coagulation, sedimentation, filtration, disinfection with ozone, and post-disinfection with chlorine). The main difference between the water treatment procedures of the two utilities is that Quebec City uses pre-chlorination of raw water while Sainte-Foy uses pre-ozonation.

3.1. DATA COLLECTION

For each utility, eighteen samples of post-ozonated water were chlorinated from Feb. 2001 to Jan. 2002. Once each sample was collected, it was then subjected to bench-chlorination with 2.5 mg/L dose (using sodium hypo-chlorite). This represents the maximum post-chlorination dose applied in both utilities. The water temperature measured in the field was reproduced in the laboratory and the pH was set to 7.5 (the average value found in distribution systems for both utilities). Once chlorine was applied, THMs and HAAs were measured following six different contact times varying from 15 min to 1, 2, 6, 24 and finally to 48 hours. These contact times represent theoretical residence times of water in the distribution systems of both utilities.

THMs and HAAs were analysed according to US EPA methods 551.1 and 552.2, respectively (USEPA, 1990; USEPA, 1995). Samples for THMs and HAAs analysis were extracted using pentane and methyl-tert-butyl-ether, respectively. After sample extraction, analysis for THMs and HAAs was conducted by means of two Perkin Elmer auto-system XL gas chromatographs with electron capture detectors (Rodriguez and Sérodes, 2001; Sérodes *et al*., 2003). For THM species, analytical protocols ensured detection limits of 0.5 µg/L for TCM and of 0.3 µg/L for BDCM, DBCM, and TBM. Detection limits for DCAA and TCAA were 1.1 and

0.6, respectively. The concentrations of other HAAs were always below the detection limits, so they were not included in this analysis.

Table 6 presents the statistical portrait of the chlorinated DBPs identified according to the experimental protocol. If the values were found below detection limits, half of the detection limit values was assumed in the analysis for selected compounds. As observed, mostly non-brominated DBPs were identified in the waters of the utilities under study. This may be explained with the help of low bromide levels in the source water (<10 μg/L), which is typical of most Canadian inland waters (Health Canada, 1995). In the waters of both utilities, concentrations of THMs and HAAs increased with contact time. However, the levels of most species stabilized after 24 hours of contact.

Increasing contact times assure more DBPs concentrations in the water distribution systems. The data are reported with respect to season and chlorine contact time. The contact time is used as a surrogate for water residence time in the distribution system, which shows spatial variability in DBP concentration.

3.2. FUZZY ANALYSIS

The application of the fuzzy synthetic evaluation procedure allowed estimation of the potential risks related to DBPs in waters from the two utilities. Both DBP cancer and non-cancer risks were evaluated. Figures 2 and 3 represents seasonal variations of risk memberships (*low*, *medium* and *high*) for Quebec City and Sainte-Foy, respectively. Figure 2 represents risk memberships for 0.25, 2, 6 and 24 hour contact times for Quebec City water samples. It can be noticed that membership in *low risk* decreases as the contact time increases. But at the same time the memberships to *medium* and *high risk* increase. Seasonal effects are also very prominent; as the temperature increases (summer season), the memberships in *low risk* decreases and in *medium* and *high risk* increases. Such seasonal effects appear more significant for high contact times. Results concerning seasonal and contact time variations are similar for the Sainte-Foy water (see Figure 3). However, the memberships in *low risk* and *high risk* for Sainte-Foy are much higher and much lower, respectively, than Quebec water (see Table 7). These results suggest that, although the average concentrations of THMs and HAAs are lower than regulated levels (Table 2), the potential risk that related to these DBPs is higher in Quebec City than in

Sainte-Foy water. This result can be explained by the strategies used by these two utilities for raw water pre-oxidation. Thus, the use of pre-ozonation in Sainte-Foy would reduce significantly the potential of cancer and non-cancer risks related to chlorinated-DBPs in the distribution system.

The risk memberships were subsequently used to evaluate the risk indices using equation 8. The US EPA levels for total THMs (80 μg/L) and HAAs (60 μg/L) were then used to estimate the ratio of DBPs concentrations with respect to their threshold levels and the maximum value of the ratio (standard method value) was taken as a representative estimate. The risk indices for Quebec City data were then compared with standard method ratios to develop a predictive model. A simple linear regression model was developed, and the equation is given below. The model has an R^2 value of 0.76 (Figure 4).

$$Std.\,Method = 1.93(RI) - 0.93 \qquad (11)$$

The above model was used to predict the standard method ratios for Sainte-Foy. Figure 5 shows the estimated ratios for the standard method (based on data collected) and the ratios predicted using the above model. A high value of R^2 (0.82) was observed for the data, which indicates the good predictive capability of this model.

3.3. SENSITIVITY ANALYSIS

Sensitivity analysis is the process of estimating the degree to which outputs of a model change as the values of input parameters are changed. As mentioned before that the standard method (given in equation 9) ratio exceeding "1" means violation of regulated guidelines. Figure 6 shows the sensitivity analysis results with respect to variability in TCM concentrations. The concentrations for other five species are fixed at 10 μg/L in this example. As the concentration of TCM approaches to 50 μg/L (remaining 3 THM species have total concentration of 30 μg/L), the standard method ratio becomes steady at 1.0, but a gradual changes in memberships of *low*, *medium* and *high* risks, and risk index (*RI*) can be noted. Another important point is that though the total concentration of two HAAs species is 20 μg/L, but it does not play any role in standard ratio method, whereas the proposed method fully takes their contribution towards final risk memberships. Similar type of sensitivity analysis can be performed for other DBPs.

4. DISCUSSION

In aggregation or grouping process, recognition of two potential pitfalls namely *exaggeration* and *eclipsing* is important. Exaggeration occurs when all basic items are of relatively low risk, yet the final risk comes out unacceptably high. Eclipsing is the opposite phenomenon, where one or more of the risk items is of relatively high risk, yet the estimated aggregative risk comes out as unacceptably low. These phenomena are typically affected by the aggregation method used, thus the challenge is to determine the best aggregation method which will simultaneously reduce both exaggeration and eclipsing (Ott, 1978).

Aggregation operators used for the development of environmental indices generally include additive forms (simple addition, arithmetic average, weighted average), root sum power, root sum square, maximum, multiplicative forms (e.g., geometric mean, weighted product), and minimum operators (Silvert, 2000; Somlikova and Wachowiak, 2001; Ott, 1978).

The proposed approach has several advantages:

- It enables the synthesis of both cancer and non-cancer risk data into a single framework;
- It assigns memberships to various levels of risk instead of a crisp number (like threshold value of guidelines) and propagates that vagueness throughout the grouping process;
- Its modular form is scalable; enabling the accommodation of new knowledge and information, such as more species of DBPs and other water quality indicators;
- It can be used to guide decision-making to focus attention on those areas which have the most adverse impact on final risk;
- More toxicity data will increase the reliability of this method and can also help pinpoint those areas in which more data would yield the highest benefits; and
- It is easily programmable for computer applications and can become a risk analysis tool for a water distribution system;

The limitations of the proposed method are:

- It may be sensitive for the selection of weights and aggregation operators. A trial and error approach may be required to avoid exaggeration and eclipsing;

- No interaction effects (synergistic or antagonistic) are considered while developing risk indices and various species of DBPs are assumed to have independent effects; and

- This framework accommodates both highly reliable and less reliable data. Some toxicity data may be supported by rigorous observations, while other data may be based on extrapolation models. These two types of data should have different weights in the aggregation process. The structure in its current form does not address this need to distinguish between data obtained from sources of different reliabilities.

The structure presented in this paper is a simplified demonstration of the approach. A comprehensive structure would require a major effort, including the collaboration of several experts in the various disciplines of water quality.

5. SUMMARY AND CONCLUSIONS

The water in distribution networks may contain various types of DBPs, which are harmful for human health. The quantification and characterization of risk in water distribution systems is a complex process. In this study, a risk-index was developed using cancer and non-cancer risk data of THMs and HAAs. A fuzzy synthetic evaluation technique is applied for aggregating risk posed by various DBPs species. An analytic hierarchy process was used for the aggregation of the risk items. Weighted average operators were used for grouping various risk items that may be expressed in non-commensurate units. The selection of appropriate aggregation operators can be challenging. Future research should develop a comprehensive system, including expert panels, and processes for the selection of the most appropriate aggregation operators.

Future research must also consider other chlorinated-DBPs (e.g., acetonitriles) and non-chlorinated DBPs occurring in drinking water such as those produced by disinfection with ozone (i.e., bromates) and with chlorine dioxide (i.e., chlorite and chlorate). Indeed, several water utilities (even those presented in this study) use more than one type of disinfectant. Thus, an improved risk-index must integrate the risk related to non-chlorinated DBPs. In addition, future

work must also be carried out to establish a unified risk-index by incorporating potential infectious risk associated with microbiological water quality.

In the model development stages, the risk-index is expected to have limited meaning for the acceptability of risk by public. It is envisaged that as this model is developed, populated and subsequently improved upon (using newly obtained data) the developers will gain insight into acceptable risk levels as they are manifest in the final fuzzy and/or defuzzified risk values. In the longer term, this approach could serve as a basis for bench marking acceptable risk due to DBPs in water distribution system.

As shown that a good correlation exists between standard method and the proposed risk index, it can be concluded that what can be achieved through former, can be predicted by proposed approach (meaning whether meeting regulatory requirements or not). In addition, the proposed approach also provides the memberships to various risk levels, which can not be inferred from standard US EPA method. The evaluation of risk, based on the fuzzy-synthetic evaluation procedure presented in this paper, can be applied by different users: water managers (for example for selecting between water treatment procedures according to the DBP risk reduction), government managers responsible for regulations (e.g., as a decision-making tool to regulate specific DBPs) and environmental epidemiologists (e.g., to estimate human exposure to DBPs in drinking water for cancer and non-cancer epidemiological studies).

The application presented here used fuzzy synthetic evaluation for risk indexing of chlorinated DBPs which were generated by laboratory-scale studies (that is, potential DBP occurrence). Once available, the method must be also validated in the future with robust full-scale data representing seasonal and spatial variations of DBPs in distribution systems.

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Table 1. Toxicological information on DBPs (modified after US EPA, 1999)

DBPs	Compound	Detrimental effects
Trihalomethanes (THMs)	Chloroform	Cancer, liver, kidney, and reproductive effects
	Dibromochloromethane	Nervous system, liver, kidney, and reproductive effects
	Bromodichloromethane	Cancer, liver, kidney, and reproductive effects
	Bromoform	Cancer, nervous system, liver and kidney effects
Haloacetic acids (HAAs)	Dichloroacetic acid	Cancer, reproductive and developmental effects
	Trichloroacetic acid	Liver, kidney, spleen and developmental effects

Table 2. DBPs (in μg/L) regulations in various jurisdictions of the world

Compound	Acronym	WHO (1993)	US EPA (2001)	Canada (2001)	Aus-NZ (2000)	UK (2000)
Trichloromethane (chloroform)	TCM	200	0[2]			
Dibromochloromethane	DBCM	100	0[2]			
Bromodichloromethane	BDCM	60	60[2]			
Tribromomethane (bromoform)	TBM	100	0[2]			
Total trihalomethanes	TTHM	$\sum_{i=1}^{4} \frac{THM}{WHO} \leq 1$	80	100	250	100
Monochloroacetic acid	MCAA				150	
Dichloroacetic acid	DCAA	50			100	
Trichloroacetic acid	TCAA	100			100	
Haloacetic acids	HAA5		60	[1]		

1: Under consideration

2: Maximum contaminant level goals (MCLG)

Table 3. Carcinogenic and non-cancer risk information for selected DBPs

Disinfection by-products (DBPs)	Carcinogenic potential	[1]Risk (1×10^{-6}) (μg/L)	[2]UF	[3]MF	RfD (mg/kg/day)	[4]RQ (μg/L)
† Chloroform	Probable (B2)	[5]5.7	10^3	1	1×10^{-2}	350
† Dibromochloromethane	Possible (C)	0.4	10^3	1	2×10^{-2}	700
† Bromodichloromethane	[6]Not classifiable (D)	[8]0.6			No data	[7]60
† Bromoform	Probable (B2)	4	10^3	1	2×10^{-2}	700
*Monochloroacetic acid			[9]10^5		No data	91
*Dichloroacetic acid	Probable (B2)	[8]0.5			No data	[10]50
*Trichloroacetic acid	Possible (C)	[8]1			No data	[10]100
*Monobromoacetic acid			[11]10^5		No data	62
*Dibromoacetic acid			[12]10^5		No data	610
Bromochloroacetic acid			[13]10^3	1	No data	1400

1: Concentration corresponding to unit risk of 1 in a million

2: Uncertainty factor; 3: Modifying factors - these are factors used to convert NOAEL and LOAEL into RfD

4: Concentration estimated at risk quotient (RQ) = 1;

where RQ = Dosage/RfD and Dosage = [Concentration × intake rate/body weight]

5: Estimated from slope factor (SF) of 0.0061 $(mg/kg/day)^{-1}$; where Risk = Dosage × SF

6: Also classified as B2 (see IRIS, US EPA, 2003)

7: US EPA (2001) and WHO (1993)

8: Derived from RQ/100

9: LD_{50} = 260 mg/kg/day (Berardi *et al*., 1987), the UF = 10^5 is assumed to estimate conc. corresponding to RQ = 1

10: WHO (1993)

11: LD_{50} = 177 mg/kg/day (Linder *et al*., 1994), the UF = 10^5 is assumed to estimate conc. corresponding to RQ = 1

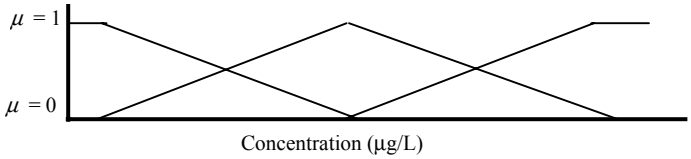
12: LD_{50} = 1737 mg/kg/day (Linder *et al*., 1994), the UF = 10^5 is assumed to estimate conc. corresponding to RQ = 1

13: NOAEL = 41 mg/kg/day (NTP, 1998), the UF = 10^3 is assumed to estimate conc. corresponding to RQ = 1

† : Included in THMs (a group of 4 trihalomethanes, the US EPA (2001) recommended the value of 80 μg/L)

*: Included in HAA5 (a group of 5 haloacetic acids, the US EPA (2001) recommended the value of 60 μg/L)

Table 4. Triangular fuzzy numbers for cancer and non-cancer risks

	Compounds	$^{1}\mu_L$	$^{2}\mu_M$	$^{3}\mu_H$
				
Cancer	TCM	[<5.7, 5.7, 57]	[5.7, 57, 570]	[57, 570, >570]
	DBCM	[<0.4, 0.4, 4]	[0.4, 4, 40]	[4, 40, >40]
	BDCM	[<0.6, 0.6, 6]	[0.6, 6, 60]	[6, 60, >60]
	TBM	[<4, 4, 40]	[4, 40, 400]	[40, 400, >400]
	DCAA	[<0.5, 0.5, 5]	[0.5, 5, 50]	[5, 50, >50]
	TCAA	[<1, 1, 10]	[1, 10, 100]	[10, 100, >100]
Non-cancer	TCM	[<3.5, 3.5, 35]	[3.5, 35, 350]	[35, 350, >350]
	DBCM	[<7, 7, 70]	[7, 70, 700]	[70, 700, >700]
	BDCM	[<0.6, 0.6, 6]	[0.6, 6, 60]	[6, 60, >60]
	TBM	[<7, 7, 70]	[7, 70, 700]	[70, 700, >700]
	DCAA	[<0.5, 0.5, 5]	[0.5, 5, 50]	[5, 50, >50]
	TCAA	[<1, 1, 10]	[1, 10, 100]	[10, 100, >100]

[1][value with respect to membership 1, value with respect to membership 1, value with respect to membership 0]

[2][value with respect to membership 0, value with respect to membership 1, value with respect to membership 0]

[3][value with respect to membership 0, value with respect to membership 1, value with respect to membership 1]

Table 5. Weights estimated by AHP for grouping of DBPs

Weights	Cancer	Non-cancer
w_{TCM}	0.231	0.226
w_{DBCM}	0.115	0.226
w_{BDCM}	0.077	0.172
w_{TBM}	0.231	0.226
w_{DCAA}	0.231	0.075
w_{TCAA}	0.115	0.075
Weights	Risk type	
w_{cancer}	0.67	
$w_{non\text{-}cancer}$	0.33	

Table 6. Statistical portrait (average and standard deviation of concentration are shown, μg/L) for DBPs species for waters of the two utilities for around the year sampling

Location	Contact time (hr)	DCAA	TCAA	TCM	DBCM	BDCM	TBM
Quebec City	0.25	13.7 (9.9)*	11.4 (7.9)	19.3 (15.6)	1.1 (2.2)	1.7 (1.2)	n.a**
	1	12.7 (10.5)	10.5 (9.5)	24.8 (18.3)	0.7 (1.7)	2.1 (1.5)	n.a
	2	10.9 (10)	9.2 (9.2)	26.6 (18.6)	0.6 (1)	2.4 (1.2)	n.a
	6	17.7 (13)	15.6 (12.1)	30.7 (24.4)	1.1 (2.1)	2.6 (1.6)	n.a
	24	25.1 (11)	22.8 (12)	32.0 (26.1)	1.1 (1.7)	2.6 (1.6)	n.a
	48	25.0 (13.8)	22.9 (14.9)	39.4 (34.8)	0.8 (1)	2.6 (1.6)	n.a
	Overall	17.5 (12.6)	15.4 (12.3)	28.8 (24.1)	0.9 (1.6)	2.3 (1.5)	n.a
Sainte-Foy	0.25	1.5 (1.8)	1.6 (0.7)	4.3 (2.2)	2.0 (1.3)	1.9 (1)	0.0 (0.1)
	1	2.0 (1.8)	1.8 (0.4)	5.7 (2.9)	2.4 (1.5)	3.0 (1.1)	0.1 (0.2)
	2	2.4 (1.7)	1.9 (0.8)	6.1 (4.1)	3.3 (1.9)	3.2 (1.6)	0.1 (0.2)
	6	3.2 (1.7)	2.3 (1.3)	8.3 (4.9)	3.3 (1.7)	4.5 (1.9)	0.2 (0.4)
	24	6.8 (2.7)	5.3 (2.2)	11.7 (6.1)	3.7 (2.7)	5.5 (2.8)	0.2 (0.4)
	48	6.4 (5.4)	5.4 (4.5)	17.9 (10)	3.6 (2.2)	6.7 (3.2)	0.2 (0.4)
	Overall	3.7 (3.5)	3.0 (2.7)	9.0 (7.2)	3.1 (2)	4.1 (2.6)	0.1 (0.3)

* Values in the parenthesis show standard deviation of 18 samples collected over a year for each contact time in the waters of the two utilities

** Values always below the detection limit

Table 7. Statistics of risk memberships for Quebec City and Sainte-Foy waters

Location	Memberships	Mean	Stdev.
Quebec City	r^L	0.58	0.18
	r^M	0.34	0.15
	r^H	0.08	0.07
Sainte-Foy	r^L	0.73	0.15
	r^M	0.26	0.14
	r^H	0.01	0.01

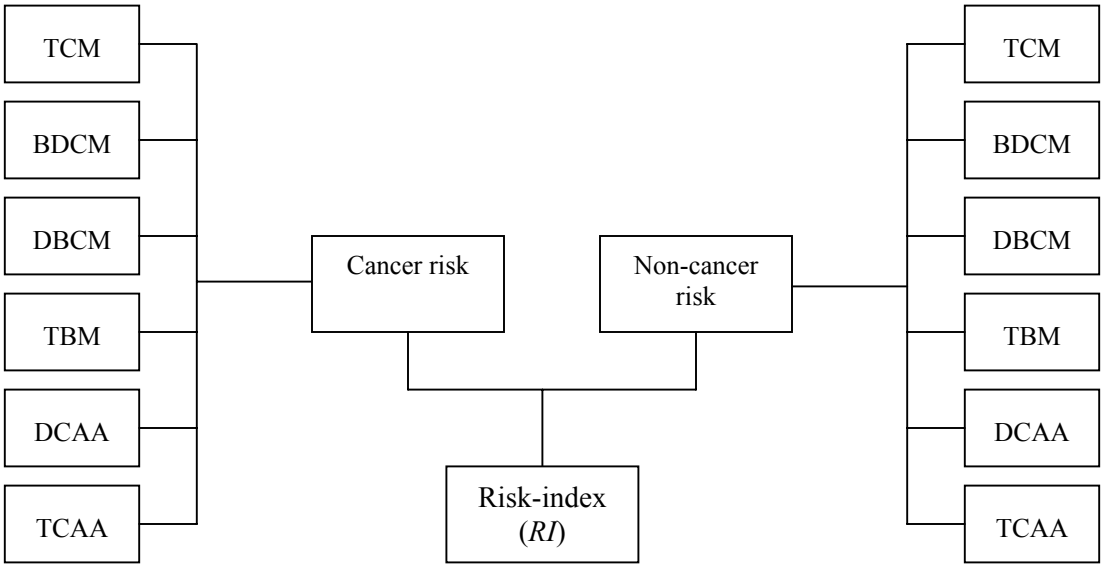
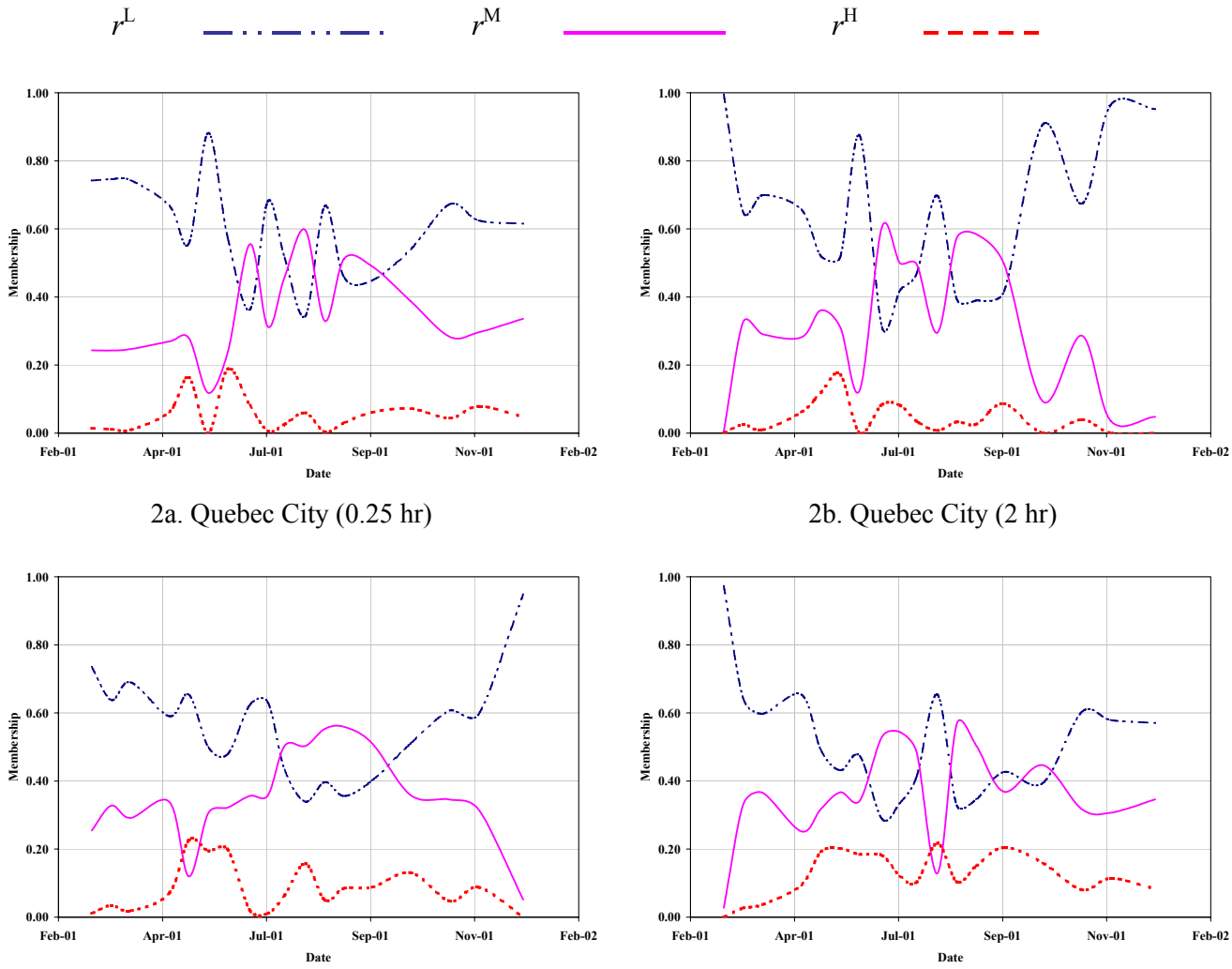


Figure 1. Grouping of various species of DBPs for their associated risk



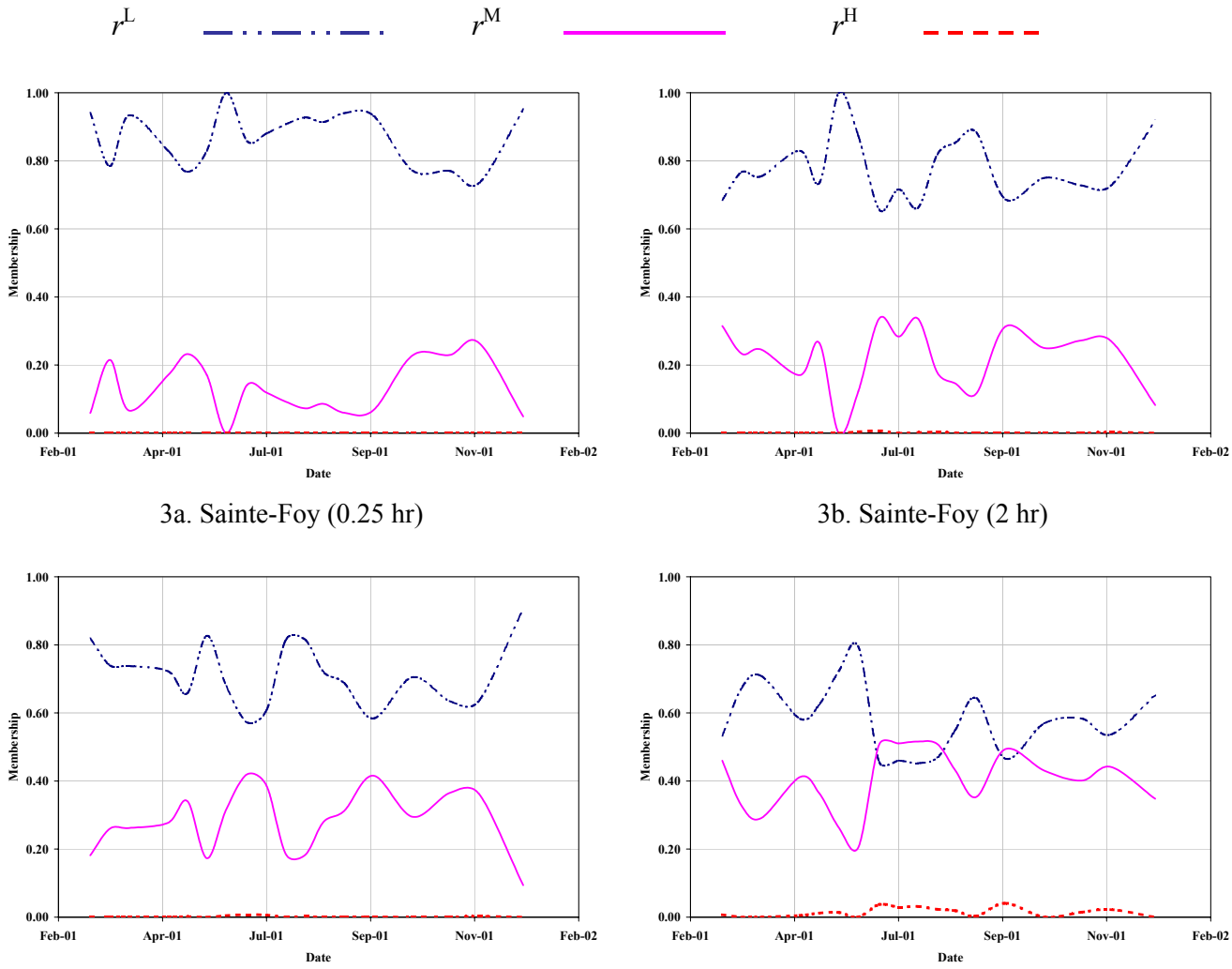
2a. Quebec City (0.25 hr)

2b. Quebec City (2 hr)

2c. Quebec City (6 hr)

2d. Quebec City (24 hr)

Figure 2. Seasonal variation of memberships of risk for Quebec City waters



3a. Sainte-Foy (0.25 hr)

3b. Sainte-Foy (2 hr)

3c. Sainte-Foy (6 hr)

3d. Sainte-Foy (24 hr)

Figure 3. Seasonal variation of memberships of risk for Sainte-Foy waters

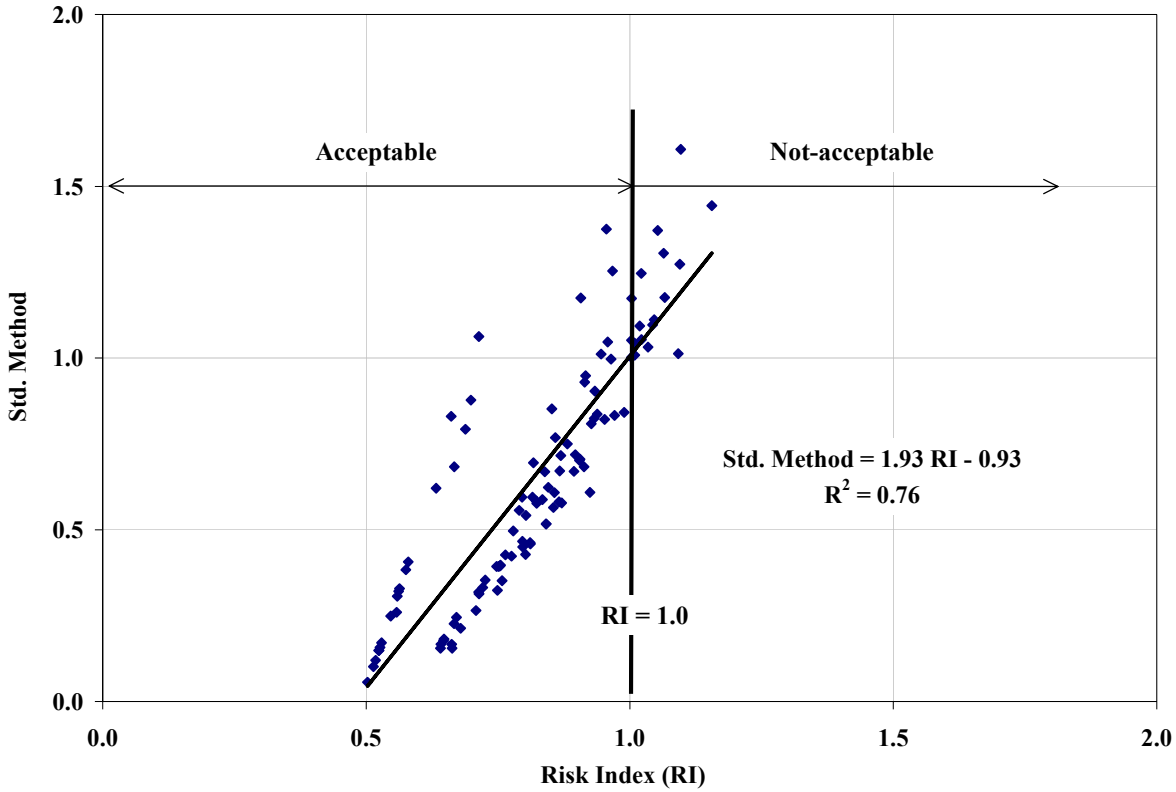


Figure 4. Comparison of risk-index with standard method based on US EPA regulations (Quebec City)

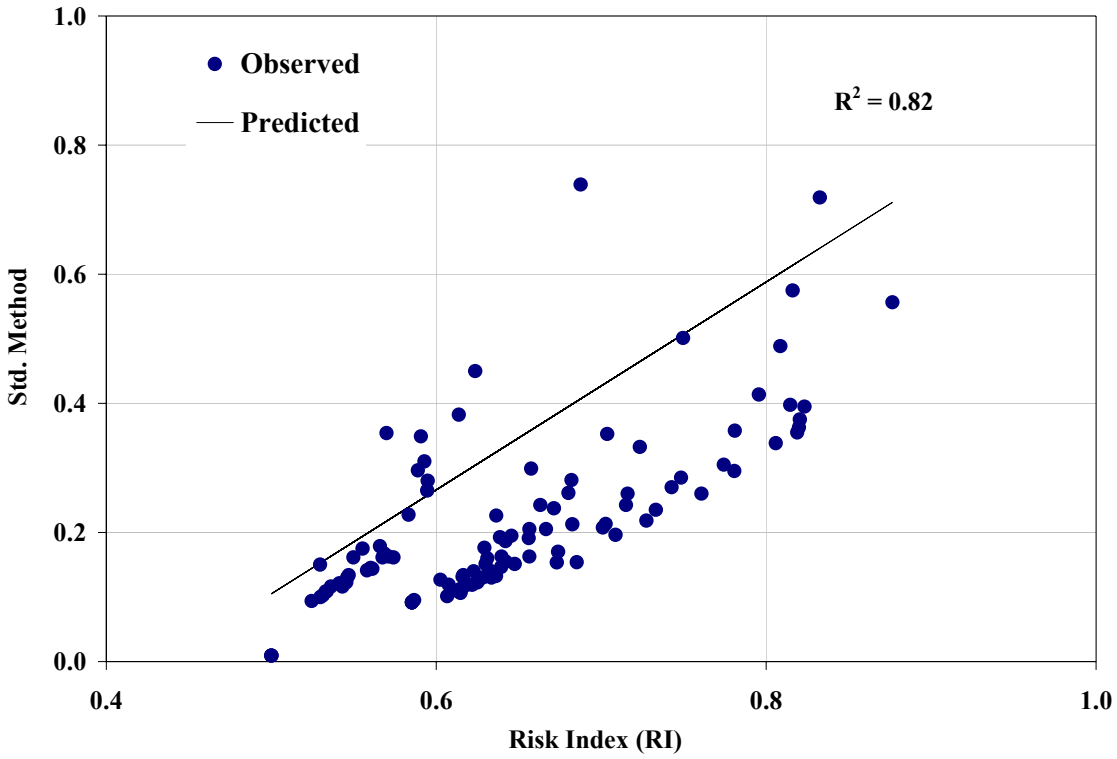


Figure 5. Comparison of risk-index with standard method based on US EPA regulations (Sainte-Foy)

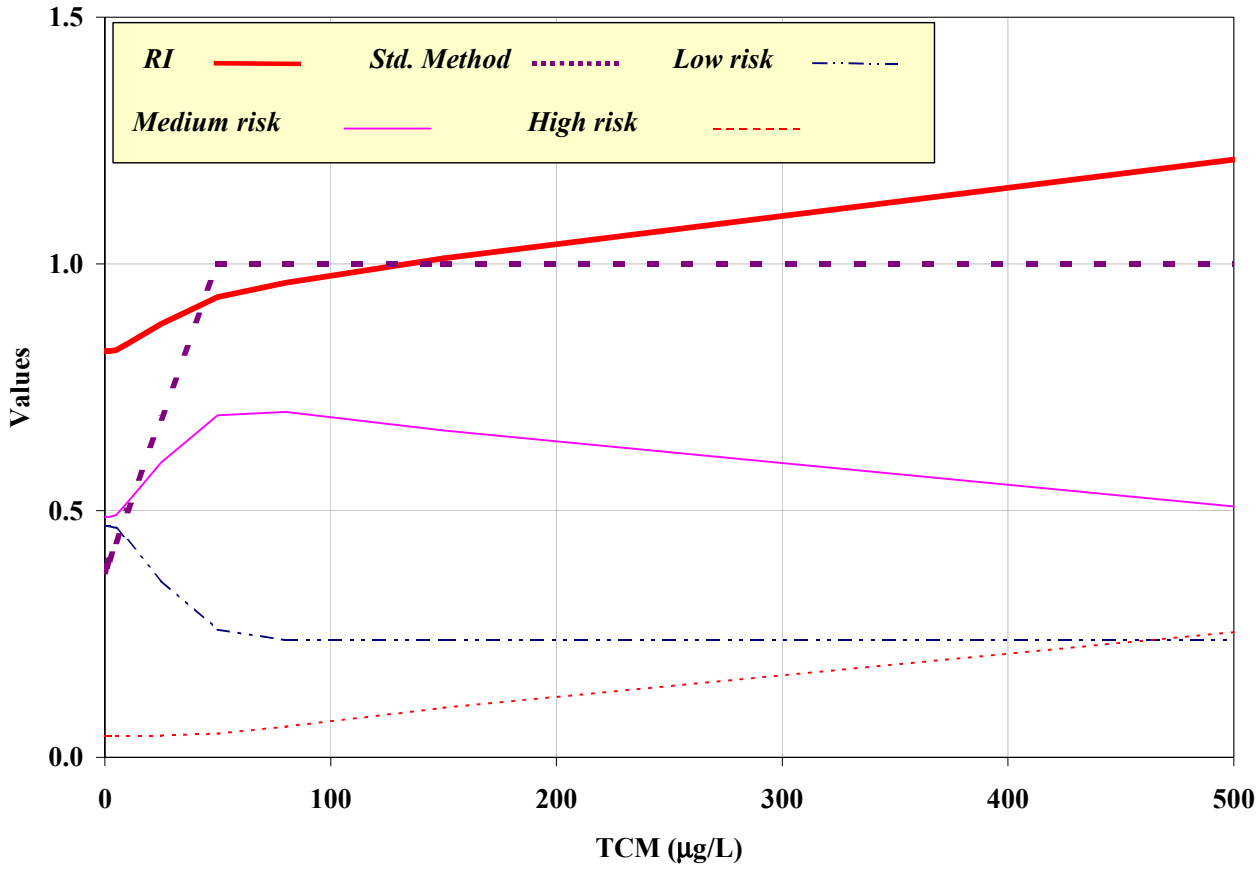


Figure 6. Sensitivity analysis of the model