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Chapter 7:
Trihalomethane Formation Potential Modeling

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7 Trihalomethane Formation Potential Modeling

[Editor's Note: This is an electronic reprint of the original document. Electronic copies of the original figures were not available, thus the original figures are not included in this report.]

The Sacramento-San Joaquin Delta is a source of drinking water for 20 million Californians. Because the Delta is part of a tidal estuary and its land use is predominantly agricultural, Delta waters tend to reflect high levels of bromides and organic material. Organics and bromides promote the formation of disinfection by-products (DBPs) in the presence of a strong oxidant. Trihalomethanes (THMs), one class of DBPs, are a suspected threat to human health when present in sufficient quantities in drinking water.

In 1979, the Environmental Protection Agency (EPA) established a drinking water standard of 0.1 milligrams per liter for THMs. Anticipating revisions to the current standards and recognizing problems Delta water users may face in meeting more stringent requirements, the Department of Water Resources (DWR) has been studying THM precursors in Delta waters for several years. More recently, the Department has become active in modeling THM precursor fate and movement in the Delta as well as modeling THM formation and speciation.

This chapter summarizes DWR's most recent efforts to mathematically model THM formation potential in Delta waters. The first section presents a comparison between DWR's model and the THM kinetic equations employed by the EPA-Malcolm Pirnie (MPI) water treatment plant (WTP) model (Harrington et al. 1992). The second section of this chapter summarizes work that was undertaken to characterize THM speciation according to first order chemical kinetics. The final section briefly discusses a current project to simulate historically-observed THM precursor transport in the Delta.

A Comparison with EPA-MPI THM Equations

At the request of the Municipal Water Quality Investigations committee, a comparison of the modeling approaches developed by DWR (Hutton and Chung 1992a, 1992b, 1993a, 1993b) and EPA-MPI (Harrington et al. 1992) was undertaken. The DWR approach was contrasted with EPA-MPI's THM kinetic equations, not with the entire WTP model. Several aspects of the EPA-MPI THM equations were evaluated both qualitatively and quantitatively. Sensitivity analyses show the EPA-MPI kinetic equations tend to respond erroneously (in an incremental sense as well as in an absolute sense) to changes in bromide concentration.

To provide an "apples-to-apples" comparison, the DWR model was recalibrated with the University of Arizona database (Amy et al. 1987) to generalize its applicability to varying reaction conditions. This database is herein referred to as the UA database. Although the recalibrated DWR model is similar to the EPA-MPI model in terms of fit to the UA database, the DWR model responds in a more appropriate manner to incremental changes in bromide concentration. Furthermore, the DWR model requires the calibration of only two equations (14 model constants) to predict individual THM species concentrations, compared with EPA-MPI requiring calibration of 6 equations (38 model constants).

Recognizing limitations associated with high chlorine doses used to develop the UA database, the EPA-MPI and DWR models were recalibrated with a small data set provided by the Metropolitan Water District (MWD). This exercise showed the DWR formulation provides a superior fit to observed data and provides superior sensitivity to incremental changes in bromide concentration. This exercise also illustrated the potential difficulty of calibrating the EPA-MPI formulation to a small database.

Conclusions and Recommendations

Based on the aforementioned comparison between THM models, the following recommendations are offered:

1. An extensive database being developed by MWD should be used to calibrate a THM submodel employing the bromine distribution factors, rather than relying on the current EPA-MPI power function formulation.

2. It may be desirable to include different variables or otherwise improve upon the proposed form of the bromine incorporation factor (η). For example, Symons et al. (1993) shows that the initial bromide to average free available chlorine molar ratio is an important variable in predicting η. The DWR approach is not constrained to predicting η directly, however. As an alternative to predicting η directly, chloroform concentration could be predicted, possibly in a manner similar to the current EPA-MPI methodology. Then from the bromine distribution factor relationship $s_0 = [CHCl_3]/[TTHM]$, s_0 could be estimated. And since s_0 is functionally related to η, the remaining THM species could be determined. This may be an attractive alternative, particularly if differential rate equations can be developed for chloroform and [TTHM].

3. The bromine distribution factor relationships have been validated for delta waters treated under a variety of conditions. They have also been shown to be valid for the waters and conditions incorporated in the UA database. Nevertheless, these equations should be validated with additional data to test their general applicability. Assuming the worst case in which the bromine distribution factors have to be recalibrated

for different waters, the proposed formulation will require the calibration of six equations with 26 constants (which is still preferable to the EPA-MPI requirement of six equations and 38 model constants).

4. For planning studies that focus on source water management impacts, it may be desirable to agree upon a set (or sets) of "standard" treatment conditions, e.g. simulated distribution system (SDS-THM) three-hour or 24-hour reaction conditions. These conditions can then be substituted into the THM submodel to develop a simplified form that varies only with influent water quality conditions. This simplified set of equations can be used to estimate THM formation from DWRDSM model output. This simplified analysis of source water management impacts is referred to as a "Level I" analysis.

5. Similar to recommendation 4, for planning studies that focus on WTP design and operational impacts, it may be desirable to agree upon a set (or sets) of "standard" influent conditions, e.g., critical winter, dry summer, or normal year. These conditions can then be employed as input to the EPA-MPI WTP model. A simplified analysis of WTP design and operational impacts is also referred to as a "Level I" analysis.

6. Finally, more refined planning studies (particulary those where THM formation at a particular WTP is the main objective) may wish to consider source water management and WTP design and operations as one system. For this type of study, DWRDSM output can be used directly as input to the EPA-MPI WTP model. This more sophisticated approach is referred to as a "Level II" analysis.

Critique of EPA-MPI THM Equations

The EPA-MPI THM equations were evaluated both qualitatively and quantitatively. The qualitative critique focused on model form, while the quantitative critique focused on model sensitivity.

Questionable Functional Forms. Amy et al. (1987) emphasized the development of "chemically rational yet statistically valid" models. Many aspects of the EPA-MPI THM equations do not adhere to this philosophy:

1. The precursor-related parameter UVA*DOC was determined by Amy et al. to be the best overall in terms of chemical significance and statistical fit. According to the authors, "The chemical significance of this parameter is that DOC represents a means of defining precursor concentration while UV absorbance provides an indication of precursor reactivity in forming THMs". The EPA-MPI equations for individual THMs use a variety of precursor-related parameters: UVA*DOC,

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UVA/DOC, UVA, and Br/DOC. This deviation from a single precursor-related parameter appears to be a compromise of "chemical significance" for statistical fit. The EPA-MPI equations (six equations and 38 model constants) developed from the UA database are as follows:

$$CHCl_3 = 0.278 \times (UVA \times DOC)^{0.616} \times (Cl_2)^{0.391} \times (t)^{0.265} \times (T)^{1.15} \times (pH - 2.6)^{0.800} \times (Br + 1)^{-2.23} \quad \text{............} \quad (1)$$

$$CHCl_2Br = 0.863 \times (UVA \times DOC)^{0.177} \times (Cl_2)^{0.309} \times (t)^{0.271} \times (T)^{0.720} \times (pH - 2.6)^{0.925} \times (Br)^{0.722} \quad \text{..............} \quad (2)$$

$$CHClBr_2 = 2.57 \times (UVA/DOC)^{-0.184} \times (Cl_2)^{-0.0746} \times (t)^{0.252} \times (T)^{0.570} \times (pH - 2.6)^{1.35} \times (Br)^{2.06} \quad \text{................} \quad (3)$$

$$CHBr_3 = 61.4 \times (UVA)^{0.683} \times (Cl_2)^{-0.176} \times (t)^{0.110} \times (T)^{-0.0596} \times (pH - 2.6)^{1.89} \times (Br/DOC)^{1.79} \quad \text{..................} \quad (4)$$

$$[TTHM] = 0.00309 \times (UVA \times DOC)^{0.440} \times (Cl_2)^{0.409} \times (t)^{0.265} \times (T)^{1.06} \times (pH - 2.6)^{0.715} \times (Br + 1)^{0.036} \quad \text{.........} \quad (5)$$

$$AMW = 105 \times (UVA)^{-0.089} \times (Br + 1)^{0.48} \quad \text{...} \quad (6)$$

2. The EPA-MPI model does not sum the four individual THM species predictions to arrive at a total mass weight. Rather, the predictive approach is to estimate total mass weight as the product of total molar weight (Eq. 5) and apparent molecular weight (Eq. 6). This approach does not constrain total THMs to equal the sum of the four species. The magnitude of deviation associated with this approach has not been explored.

3. Eq. 6 does not take advantage of a priori knowledge of boundary conditions, i.e. a minimum AMW of 119.4 µg/µmole at 100 percent chloroform and a maximum AMW of 252.7 µg/µmole at 100 percent bromoform. Disregarding these boundary conditions permits the regression equation to predict infeasible values under extreme conditions.

4. Eqs. 1 through 4 do not approximate the nonlinear response of THM formation to bromide over a wide range of bromide. Fig. 1 gives examples of the response to bromide as observed by others. Harrington et al. (1991) attemped to circumvent this problem by segregating data into bromide ranges and modeling in a piece-wise fashion. While a piece-wise approach is certainly valid, it can result in a discontinuity at the interface between bromide ranges.

5. The resulting piece-wise equations show that while a given parameter may be directly related to THM formation under one bromide range, the same parameter may be inversely related to THM formation under another bromide range. This behavior, while easily handled by the

DWR model with the bromine distribution factors, is not addressed by Eqs. 1 through 4.

Sensitivity Analyses

Performing sensitivity analyses on the EPA-MPI individual THM equations revealed erroneous model sensitivities, particularly with respect to bromide.

1. The base conditions employed for sensitivity analyses are adopted from Chowdhury et al. (1991), a paper on the original development of the EPA-MPI individual THM equations. Base conditions are: DOC = 3 mg/L, UVA = 0.045, pH = 7.5, Cl2 = 4 mg/L, T = 25 C, and bromide takes on alternate values of 0.03, 0.3 and 0.6 mg/L. An additional base condition set for this analysis was t = 24 hrs.

2. See Figs. 2 and 3. While the CHCl3 sensitivity given by Eq. 1 appears to follow the general pattern shown in Fig. 1, the CHCl2Br and CHClBr2 estimates from Eqs. 2 and 3 "blow up" with increases in bromide. The CHBr3 estimates from Eq. 4 are relatively insensitive to bromide increases.

3. The piece-wise approach constrains the model from "blowing up". However, note the extreme discontinuities produced by this approach. The piece-wise model results in CHBr3 being even less sensitive to bromide increases and results in total THMs decreasing with increasing bromide, an erroneous result. Fig. 4 shows TTHM as the sum of Eqs. 1 through 4, rather than as the product of Eqs. 5 and 6.

Recalibration of DWR's Model

DWR's model was originally developed to predict THM formation under pre-defined test conditions, first for THMFP and later for SDS-THM. In this study, the model was reconfigured to predict individual THM compounds under varying test conditions and was calibrated with the UA database. As with the original formulation, individual species mass concentrations are calculated as follows:

$$CHCl_3 = 119.36 \times [TTHM] \times s_0(\eta) \dotfill (7)$$

$$CHCl_2Br = 163.82 \times [TTHM] \times s_1(\eta) \dotfill (8)$$

$$CHClBr_2 = 208.28 \times [TTHM] \times s_2(\eta) \dotfill (9)$$

$$CHBr_3 = 252.74 \times [TTHM] \times s_3(\eta) \dotfill (10)$$

where [TTHM] is now predicted from Eq. 5 and s_0, s_1, s_2 and s_3 are the bromine distribution factors previously defined as:

$$s_0(\eta) = -0.222\eta^3 + 0.2444\eta^2 - 0.8667\eta + 1 \quad \text{...(11)}$$

$$s_1(\eta) = 0.0731\eta^3 - 0.4621\eta^2 + 0.7288\eta \quad \text{...(12)}$$

$$s_2(\eta) = -0.0753\eta^3 + 0.1723\eta^2 + 0.1607\eta \quad \text{..(13)}$$

$$s_3(\eta) = 0.0296\eta^3 + 0.0222\eta^2 \quad \text{...(14)}$$

It was unnecessary to recalibrate Eqs. 11 through 14 to the UA database. The bromine incorporation factor (η) was modeled with a form previously suggested:

$$\eta = \frac{k}{1+\beta} \quad \text{..(15)}$$

where k is the bromine saturation level and takes on a value of 3. To predict THM speciation under varying test conditions, β was expanded into a multivariable function:

$$\beta = 0.418 \times (UVA \times DOC)^{0.551} \times (Cl_2)^{0.133} \times (t)^{0.034} \times (T)^{0.367} \times (pH - 2.6)^{-0.174} \times (Br)^{-1.07} \quad \text{.......................(16)}$$

The generalized bromine incorporation factor formulation shown in Eqs. 15 and 16 are preliminary and alternative formulations are being considered. Eq. 16 was developed with a backward stepwise log-linear regression procedure using a copy of the UA database provided by MPI. The database has 1,025 data points varying somewhat from the 995 database reported by Amy et al. (1987). As a caveat on the bromine incorporation factor, note that the EPA-MPI model implicitly uses the concept of bromine incorporation factor through the AMW term. AMW is a linear function of the bromine incorporation factor:

$$AMW = 119.36 + 44.46 \times \eta \quad \text{..(17)}$$

Comparison of the DWR Model with the EPA-MPI Model

The previous discussion shows that the DWR model requires the calibration of two equations, one for [TTHM] and one for β. Model calibration of 14 constants is required, compared with the EPA-MPI calibration requirements of 38 model constants.

Observed values from the UA database were compared with predictions from the DWR model and the EPA-MPI model. Comparisons are shown in Figs. 5 through 7 as relative frequency histograms of percent deviation, where:

$$\% \text{ Deviation} = (\text{Predicted} - \text{Observed}) / \text{Observed} * 100 \quad \text{................................(18)}$$

1. To allow for an unbiased comparison, the EPA-MPI model was also recalibrated to the available UA database. Recalibration was necessary because, while the equations for η and AMW were based on 1025

observations (Harrington et al. 1992), Eqs. 1 through 5 were developed from only 995 observations. Recalibrated equations are as follows:

$$CHCl_3 = 0.248 \times (UVA \times DOC)^{0.646} \times (Cl_2)^{0.453} \times (t)^{0.277} \times (T)^{1.19} \times (pH - 2.6)^{0.606} \times (Br + 1)^{-1.80} \quad \text{.......} (19)$$

$$CHCl_2Br = 0.782 \times (UVA \times DOC)^{0.163} \times (Cl_2)^{0.347} \times (t)^{0.253} \times (T)^{0.696} \times (pH - 2.6)^{0.709} \times (Br)^{0.611} \quad \text{........} (20)$$

$$CHClBr_2 = 0.974 \times (UVA/DOC)^{-0.445} \times (Cl_2)^{-0.106} \times (t)^{0.216} \times (T)^{0.488} \times (pH - 2.6)^{1.15} \times (Br)^{1.86} \quad \text{..........} (21)$$

$$CHBr_3 = 63.7 \times (UVA)^{0.466} \times (Cl_2)^{-0.077} \times (t)^{0.110} \times (T)^{-0.155} \times (pH - 2.6)^{2.06} \times (Br/DOC)^{1.96} \quad \text{..............} (22)$$

$$[TTHM] = 0.00309 \times (UVA \times DOC)0.462 \times (Cl_2)^{0.409} \times (t)^{0.265} \times (T)^{1.07} \times (pH - 2.6)^{0.695} \quad \text{..................} (23)$$

Note that the Br+1 term was dropped from Eq. 23. Regression gave a negative exponent for this term. The backward stepwise procedure indicated that this term and several others were not statistically significant. Other terms were not dropped from the equations for this analysis, however.

2. For CHClBr2 predictions, percent deviation is not displayed in Fig. 6 when bromide is less than 0.10 mg/L. Similarly for CHBr3, percent deviation is not displayed in Fig. 7 when bromide is less than 0.25 mg/L. These omissions are justified by observing that within these bromide ranges the species concentrations tend to take on values much less than 1 µg/L.

3. Overall, both models are similar in their abilities (or lack thereof) to match the UA database.

DWR model sensitivity to bromide is shown in Figs. 2 through 4. Unlike the EPA-MPI model, DWR's model sensitivities correspond in a relative sense to trends shown in Fig. 1.

Calibrating Site-Specific Models

While the DWR model is superior in its incremental response to bromide, it is reasonable to assume that it does not adequately predict THM formation under delta drinking water conditions because of UA database limitations sited by Harrington et al. (1992). In an attempt to overcome this limitation, the DWR model was recalibrated with a small database provided by MWD. This data represents 60 observations of June 1992 conditions at Greene's Landing, West Branch SWP water at Foothill, and East Branch SWP water at Devil Canyon.

1. The [TTHM] equation was recalibrated with a backward stepwise log-linear regression procedure. The chlorine dose term was redefined as an "available chlorine" dose by accounting for ammonia chlorine

demands. A bromide term was not included in the calibration because the data set is biased in its distribution of bromide and precursors, i.e., Greene's Landing has low bromide and low precursors while the other stations have high bromide and high precursors. Inclusion of a chlorine residual term did not appear to improve the regression:

$$[TTHM] = 0.0203 \times (UVA \times DOC)^{0.388} \times (Cl_2)^{0.323} \times (t)^{0.245} \times (T)^{0.485} \times (pH - 2.6)^{0.899} \quad \dots\dots\dots (24)$$

2. The β equation was also recalibrated. Stepwise regression eliminated terms for t, T, and pH. Again, the chlorine dose term accounts for ammonia chlorine demand:

$$\beta = 0.217 \times (UVA \times DOC)^{0.618} \times (Cl_2)^{0.713} \times (Br)^{-1.30} \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots (25)$$

3. Bromine distribution factors (Eqs. 11 through 14) were not recalibrated.

The MPI-EPA THM equations were also recalibrated with the same data. Again, a backward stepwise log-linear regression procedure was used. This procedure eliminated a number of variables from the predictive equations, pointing out a disadvantage of using an approach that requires more equations and calibration constants. It is possible that these terms would not drop out if more data were available for calibration.

Observed values were compared with predictions from the recalibrated DWR and EPA-MPI models. The recalibrated DWR model gives superior predictions for TTHM and all four compounds. The difference between models is most pronounced for total THMs, where DWR estimates are within ±10 percent for 55 of the 60 observations and the EPA-MPI estimates are within ±10 percent for only 34 of the 60 observations. Again, the DWR model shows good sensitivity to changes in bromide while the EPA-MPI model does not.

Characterizing THM Speciation with First-Order Chemical Kinetics

By assuming that bromine is not actively involved in the oxidation of precursor material and is involved only in substitution reactions, THM speciation was modeled as a consecutive irreversible three-stage reaction:

$$CHCl_3 \xrightarrow{\ k_1\ } CHCl_2Br \xrightarrow{\ k_2\ } CHClBr_2 \xrightarrow{\ k_3\ } CHBr_3$$
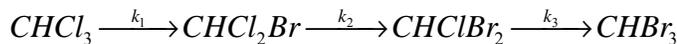
where k_1, k_2, and k_3 are first order reaction rate constants defined by the following differential equations:

$$d[CHCl_3]/d\eta = -k_1 \times [CHCl_3] \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (26)$$

$$d[CHCl2Br]/d\eta = k_1 \times [CHCl_3] - k_2 \times [CHCl_2Br] \quad \dots\dots\dots\dots\dots\dots (27)$$

$$d[CHBr_3]/d\eta = k_2 \times [CHCl_2Br] - k_3 \times [CHClBr_2]$$ ....(28)

$$d[CHBr_3]/d\eta = k_3 \times [CHClBr_2]$$ ....(29)

The following bromine distribution factor relationships result after solving this system of differential equations and assuming that the initial chloroform concentration is equal to [TTHM]:

$$s_0(\eta) = e^{-k_1\eta}$$ ....(30)

$$s_1(\eta) = \frac{k_1}{k_2 - k_1} e^{-k_1\eta} + \frac{k_1}{k_1 - k_2} e^{-k_2\eta}$$ ....(31)

$$s_2(\eta) = \frac{k_1 k_2}{(k_2 - k_1)(k_3 - k_1)} e^{-k_1\eta} + \frac{k_1 k_2}{(k_1 - k_2)(k_3 - k_2)} e^{-k_2\eta} + \frac{k_1 k_2}{(k_1 - k_3)(k_2 - k_3)} e^{-k_3\eta}$$ ....(32)

$$s_3(\eta) = 1 - \frac{k_2 k_3}{(k_2 - k_1)(k_3 - k_1)} e^{-k_1\eta} - \frac{k_1 k_3}{(k_1 - k_2)(k_3 - k_2)} e^{-k_2\eta} - \frac{k_1 k_2}{(k_1 - k_3)(k_2 - k_3)} e^{-k_3\eta}$$ ....(33)

From Eq. 30, a value for the rate constant k_1 may be estimated by plotting $\ln(1/s_0)$ versus η and determining a best-fit slope. This technique results in an estimate of $k_1=1.19$ for the calibration data. Rate constant k_2 may be estimated by setting Eq. 27 equal to zero, dividing through by [TTHM], substituting in $k_1=1.19$, and determining values of $s_0(\eta)$ and $s_1(\eta)$ for the value of η such that s_1 is maximized. This step can be satisfied by visual inspection of the data, resulting in $s_0=0.334$ and $s_1=0.340$ at $\eta=1.05$ and:

$$k_2 = 1.19 \times (0.334/0.340) = 1.17$$ ....(34)

In a similar manner, k_3 may be estimated by setting Eq. 28 equal to zero, dividing through by [TTHM], substituting in $k_2=1.17$, and determining values of $s_1(\eta)$ and $s_2(\eta)$ for the value of η such that s_2 is maximized. Values of $s_1=0.218$ and $s_2=0.411$ were estimated for $\eta=1.90$ and:

$$k_3 = 1.17 \times (0.218/0.411) = 0.62$$ ....(35)

The first order kinetic representation of the bromine distribution factors can be summarized by substituting values for the rate constants into Eqs. 30 through 33:

$$s_0(\eta) = e^{-1.19\eta}$$ ....(36)

$$s_1(\eta) = -59.50e^{-1.19\eta} + 59.50e^{-1.17\eta}$$ ....(37)

$$s_2(\eta) = 122.13e^{-1.19\eta} - 126.57e^{-1.17\eta} + 4.44e^{-0.62\eta}$$ ....(38)

$$s_3(\eta) = 1 - 63.63e^{-1.19\eta} + 67.07e^{-1.17\eta} - 4.44e^{-0.62\eta} \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (39)$$

While these relationships provide some correspondence between observed and predicted values, the correspondence is inferior to that provided by probability-based polynomial relationships (Hutton and Chung 1993b). Deviation between observed and predicted values, particularly at high values of η, may suggest that bromine substitution reactions do not always follow first order kinetics.

Historic Simulation of Delta THM Precursors

The purpose of this project is to validate DWRDSM's use as a tool to track bromide and THM precursors in the delta. DWR is currently running a 24-month DWRDSM simulation (12-month model "warm-up") of historic conditions from October 1989 through September 1991, tracking bromide, dissolved organic carbon (DOC), ultraviolet absorbance at 254 nm (UVA), and THMFP as carbon (TFPC). The simulation employs a monthly time step, a 19-year mean tide, and DAYFLOW hydrology. When available, water quality boundary conditions are based on historic grab-sample bromide and precursor data collected by DWR (Input 1993). It is anticipated that results of this historic simulation will be presented at the American Society of Civil Engineers Hydraulics Division's 1993 National Conference in San Francisco (Hutton and Enright 1993).

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