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NATURAL ORGANIC MATTER AND THE FORMATION AND CONTROL OF TRIHALOMETHANE DISINFECTION BY-PRODUCTS IN WATER

EPA THM PROJECT 2012 AND ADVICE NOTE 4 REV 2

Presentation to: EPA NOM Conference Meyrick Hotel, Galway 16th June 2016
by MICHAEL JOYCE, Water Services Director

PRESENTATION OVERVIEW

- The predominance of surface water sources in Ireland and the need for chlorination to inactivate pathogenic risk in drinking water
- The reactivity of Natural Organic Matter (NOM) as THM precursors in chlorinated water and the consequent risk of chlorinated disinfection by-product formation such as THM and HAAs
- Health risks associated with different trihalomethanes and their volatilisation to air
- The efficacy of treatment processes for THM precursor removal ahead of chlorination
- Outline of deliverables for EPA project on THM exceedances undertaken by Ryan Hanley
- Factors affecting THM formation in chlorinated supplies
- Review of site specific data on THM formation as part of EPA project
- Methodologies for mitigation of THM formation potential

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THE PREDOMINANCE OF SURFACE WATER SOURCES IN IRELAND, AND THE NEED TO REDUCE NATURAL ORGANIC MATTER (NOM) IN TREATED WATER SUPPLIES

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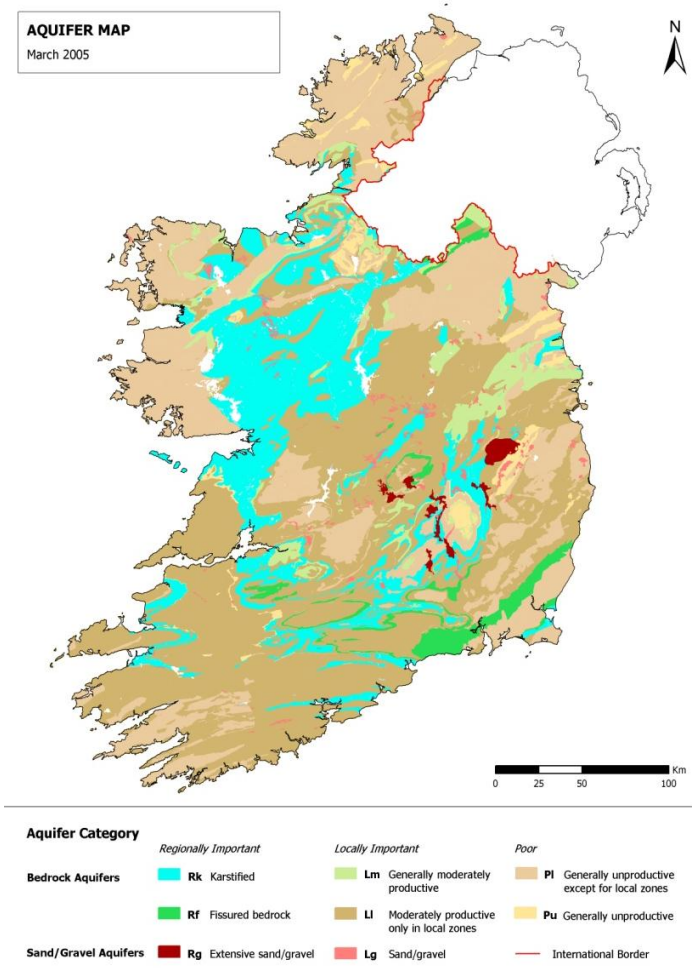
SOURCE WATER RESOURCE USE IN IRELAND



- **Ireland's public supplies have high dependence on surface waters, which is well above EU average**
  - 82% of water in Ireland is abstracted from surface water (i.e. rivers and lakes) – to approx. 50% of water supply zones
  - 18% of water from groundwater including 7.6% from springs susceptible to surface contamination
- **Surface waters particularly lowland lakes have higher level of NOM (i.e. THM precursors) than aquifer ground waters (shaded maroon) with a high risk of THM formation following chlorination if organics are inadequately removed by treatment**
- **High vulnerability also associated with ground waters sourced in karstified (i.e fissured, porous) limestone (shaded blue) overlaid by shallow overburden in the west**

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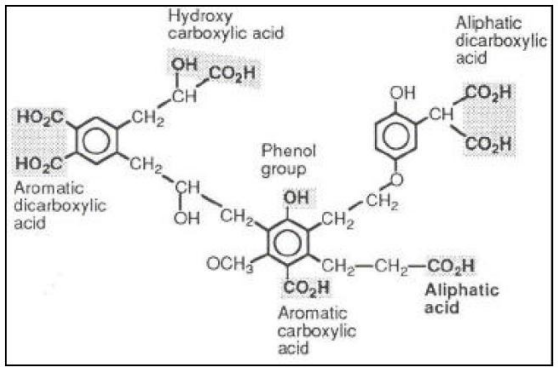
NATURAL ORGANIC MATERIAL (NOM)

- **NOM is a complex mixture of**
  - organic compounds resulting from the degradation of vegetative matter in the catchment or in the surface affected groundwater ZOC
  - Compounds resulting from the growth and decomposition of algae and weeds within the surface water source itself
- **NOM is classified into two components**
  - Humic substances namely **humic acids and fulvic acids** with molecular weights between hundreds and tens of thousands
  - Non-humic substances namely proteins, polysaccharides and carboxylic acids



Typical Humic NOM molecule

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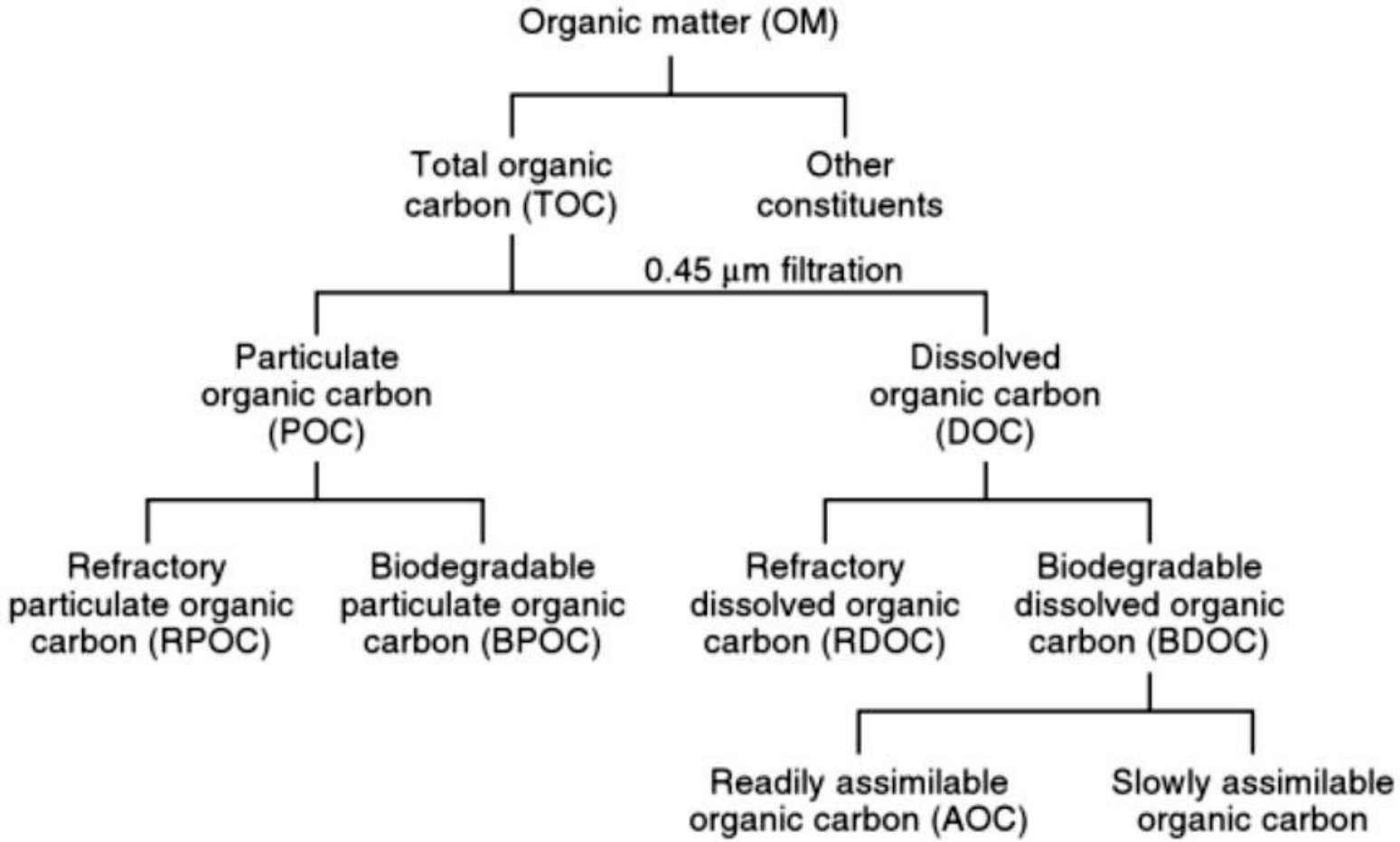
ORGANIC CARBON AS A MEASURE OF NATURAL ORGANIC MATERIAL (NOM)



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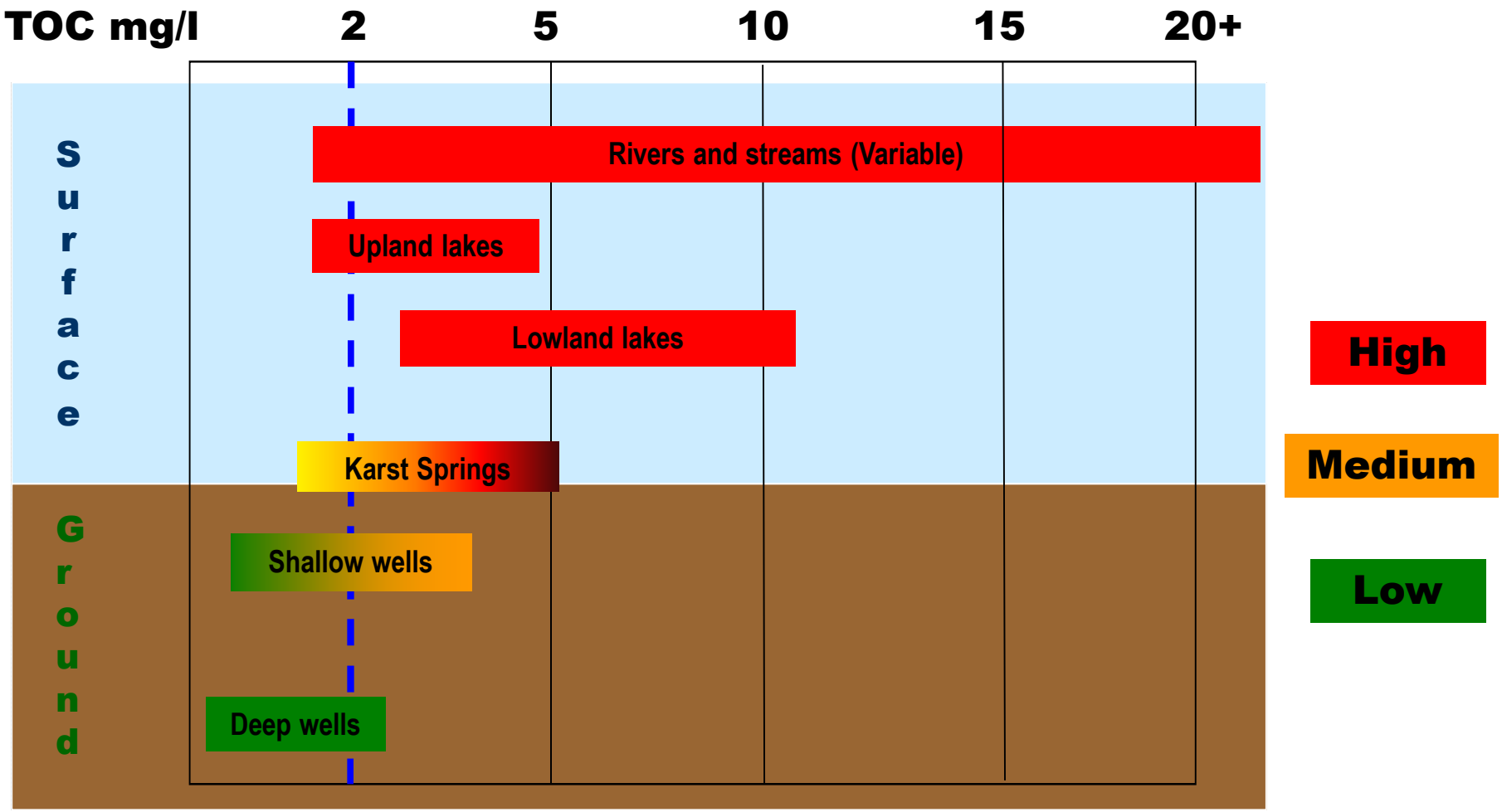
RANGE OF TOC CONCENTRATIONS IN IRISH WATERS



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THE NEED TO REMOVE NOM

- **The removal of NOM in drinking water is required because**
  - It causes colour, taste and odour in water – unacceptable to consumers;
  - It reacts with most disinfectants used in water treatment, thus reducing their disinfection efficacy in the inactivation of pathogens;
  - It influences disinfectant demand & disinfection process design and operation;
  - It can prevent the verification of UV disinfection systems
  - It affects stability and removal of inorganic particles in water;
  - It has a large effect on coagulant demand and coagulation performance;
  - It can cause irreversible fouling in membrane filtration systems;
  - It competes with taste and odour for adsorption onto GAC filters thereby reducing their adsorption capacity
  - It can affects biological re-growth in distribution systems;
  - **Chlorination of NOM produces disinfection by-products including THMs**;

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NATURAL ORGANIC MATTER (NOM) AS THM PRECURSORS IN CHLORINATED WATER SUPPLIES

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CHLORINATION FOR PRIMARY AND SECONDARY DISINFECTION

Due to the high pathogen risk associated with surface water sources or surface affected groundwater sources in Ireland, water treatment facilities in Ireland use chlorination for both primary and secondary disinfection.

PRIMARY DISINFECTION

The main disinfection method employed where a chemical or non-chemical disinfectant is used and **has to be verified** as per SI 122 of 2014 as achieving the necessary microbial inactivation of pathogenic microorganisms in water

Irish Water in their Pathogen Compliance Criteria require multi-barrier approach

a) Treatment process optimised for the attainment of pathogen removal

b) Chlorination for inactivation of bacteria and viruses using verified Ct

c) Inactivation of remaining protozoal pathogen risk using validated UV disinfection

SECONDARY DISINFECTION

The application of a chemical disinfectant primary disinfection or at some appropriate point along the distribution network to maintain the **disinfection residual** and quality assure drinking water throughout the system to the point of compliance (i.e. minimum of 0.1mg/l at the consumer tap)

Chlorination – only Monochloramine (chloramination) is possible alternative

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NOM AS THM PRECURSORS IN CHLORINATED WATER

- **Hydrophobic fraction**
  - Humic acids and lignins, which are produced from decaying vegetative matter (larger molecular weight DOC which tend to form higher levels of THMs following reaction with chlorine)
  - Lignin is resistant to biodegradation yet reactive with oxidants owing to a high density of activated aromatic chemical rings in compounds from decaying vegetative matter
  - Fulvic acids (smaller molecular weight DOC which tend to form less THMs than humic acids following reaction with chlorine)
- **Hydrophilic fraction – relatively poor THM precursors**
  - Carbohydrates and sugars (relatively poor THM precursors)
  - Compounds resulting from algal activity e.g. polysaccharides
- **Waters abstracted from different locations in a catchment and at different times of the year will have distinctly different mixtures of these THM precursors.**

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NOM REMOVAL PROCESSES

- Because of the relatively large size of dissolved humic substance molecules, their removal from water using a sufficiently tight membrane (nano-filtration)
- Because the humic substances are highly negatively charged at drinking water pH, they can be charge neutralised, coagulated, adsorbed to or enmeshed in metal hydroxide floc and removed by sedimentation or flotation followed by filtration
- Since colour is associated with aromatic content (C=C bonds) of larger humic substances, the color can be removed by breaking these bonds using a strong oxidant such as ozone
- Humic matter is generally non-biodegradable. The use of a strong oxidants like ozone can break humics into smaller, biodegradable components, removable by activated carbon or biofiltration.
- Because of this negative charge, humic matter may be removed by chemical (ion exchange) or physical adsorption (activated carbon)

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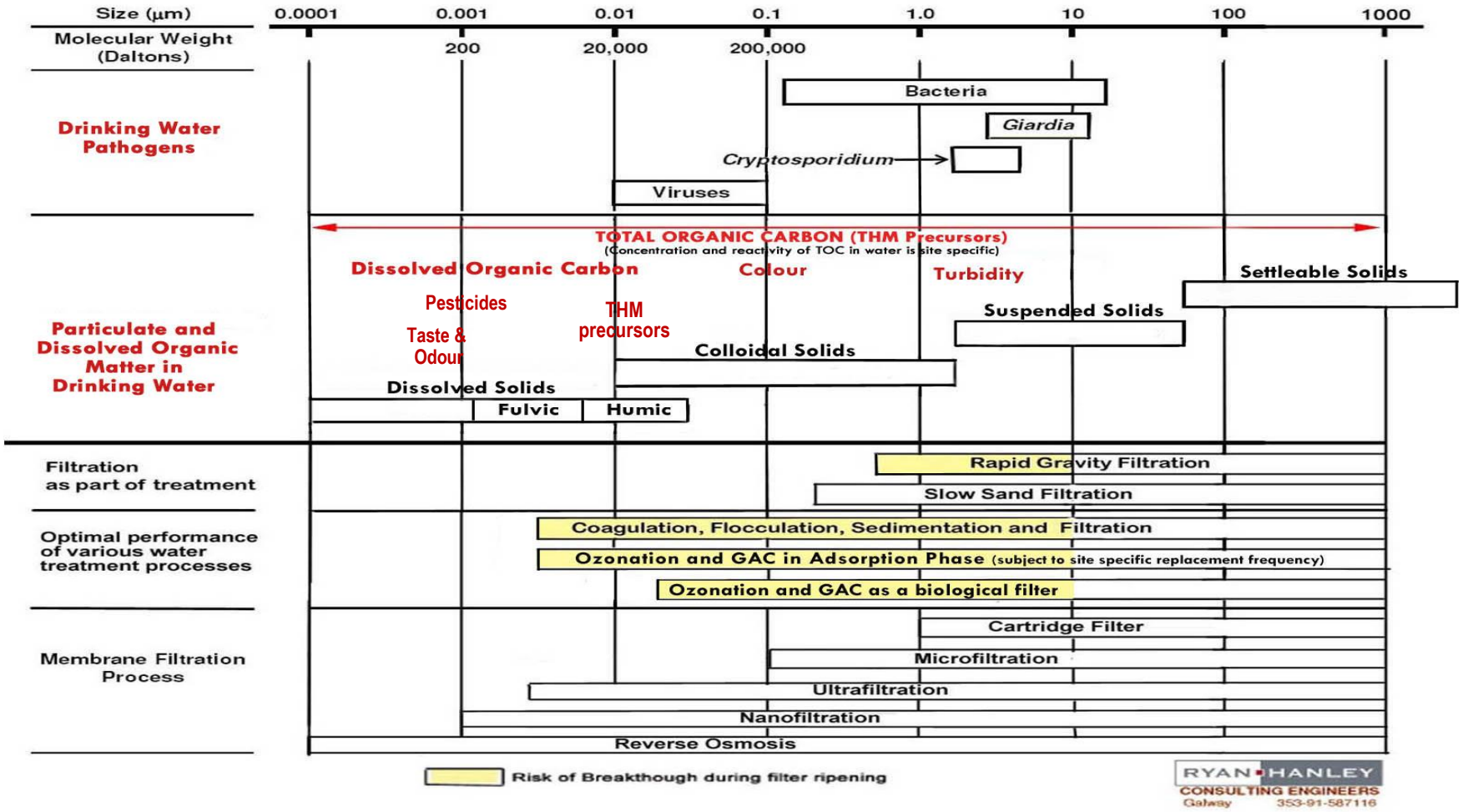
REMOVAL CAPABILITY OF WATER TREATMENT PROCESSES



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TOTAL TRIHALOMETHANES

- Cumulative total of four constituent THMs with parametric limit of 100μg/l
  - Chloroform ($CHCl_3$) - highest concentration
  - Bromoform ($CHBr_3$)
  - Dibromochloromethane ($CHClBr_2$) [DBCM]
  - Bromodichloromethane ($CHCl_2Br$) [BDCM]
- Chloroform and BDCM classified by the IARC as Group 2B compounds (i.e. possible carcinogen to humans) with chloroform linked to possible liver damage and BDCM linked to a possible increase in reproductive effects
- Bromoform and DBCM classified by the IARC as Group 3 compounds (i.e. not classifiable as carcinogen to humans)

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VOLATILISATION OF THMs

- TTHMs in water is not just a drinking water issue
- Chloroforms and BDCM are extremely volatile relative to the other two constituents of TTHMs and are ultimately transferred to air as a result of their volatility
- WHO research suggests equal contributions to total chloroforms and BDCM human exposure coming from four areas:
  - ingestion of drinking water
  - inhalation of indoor air largely due to volatilization from drinking-water
  - inhalation & dermal exposure during showering or bathing
  - ingestion of food
- Indoor air exposure is particularly related to rates of ventilation in houses and frequency of showering and bathing

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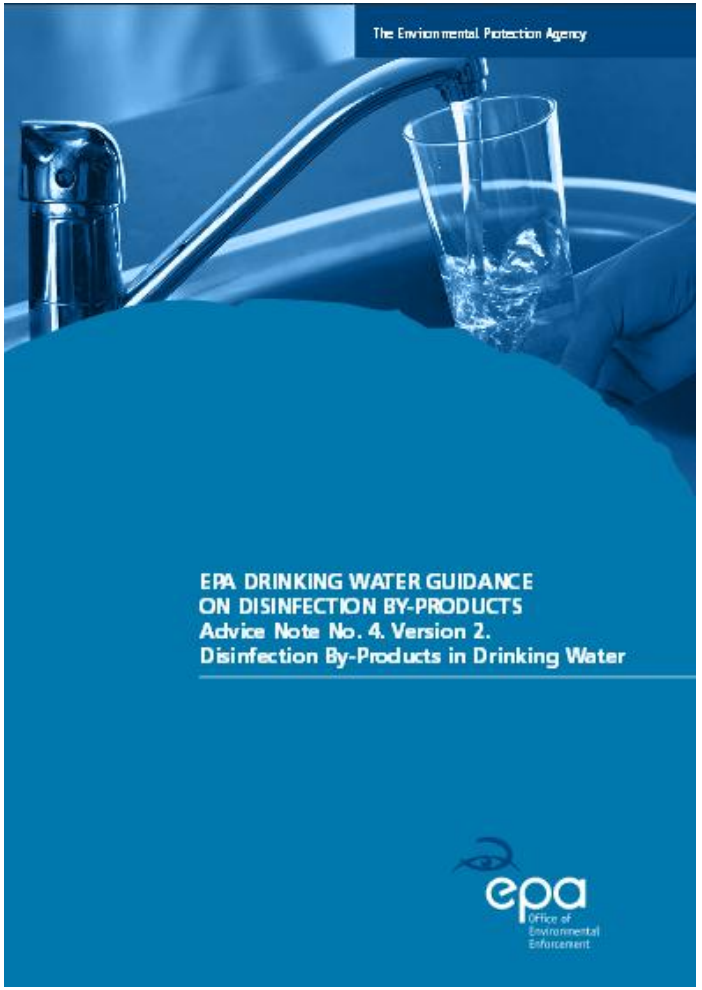
THE EPA PROJECT ON THM EXCEEDANCES

MAIN FINDINGS AND RESULTANT GUIDANCE



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EPA PROJECT - DELIVERABLES

- A desk based assessment of investigation files relating to 25 public drinking water supplies where THM exceedances have occurred and preparation of individual site reports
- Comment on the solutions put forward by the WSAs for resolving THM issues on the 25 sample supplies
- Attend the EPA site audits of 10 public drinking water supplies where frequent THM exceedances have been recorded
- Produce a technical guidance document on the overall findings of the assessment of the files and detail how these findings could be transferred to a policy or programme nationally to tackle THM exceedances at other supplies
- EPA Advice Note 4 Version - published by EPA in 2012

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FACTORS AFFECTING THM FORMATION

- Concentration of NOM in water (i.e. TOC) and the efficacy of TOC removal by treatment
- Higher chlorine dose results in higher THM potential.
- THM formation increase with increase in pH
- Rate of THM formation increase as temperature increases
- Extended contact time with chlorine.
- Sediments in distribution networks or reservoirs can react with residual chlorine.
- Ingress of surface water into reservoirs or distribution network
- Bromide concentration in water can result in brominated THMs

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THE SEASONALITY OF PEAK THM FORMATION IN DRINKING WATER

- **Seasonality in THM peaks occurs due to coincidental peaks in water temperature variation and available humic matter in early autumn**
  - In Ireland water temperature ranges from 3-18°C with the highest temperature typically recorded in late Sept/early Oct
  - THM formation in water increases with rising temperature
  - In autumn, the proportion of lignins and hydrophobic matter rises again due to the increased load of dying vegetation
  - High water temperature in the distribution system promotes the accelerated depletion of free Cl_2 residual, requiring higher doses
  - Water demands on certain schemes are often higher in summer months, resulting in lower water age helping to control THM formation during the peak summer months
  - During summer a large portion of NOM in water is hydrophilic in nature due to algal activity and less reactive with chlorine

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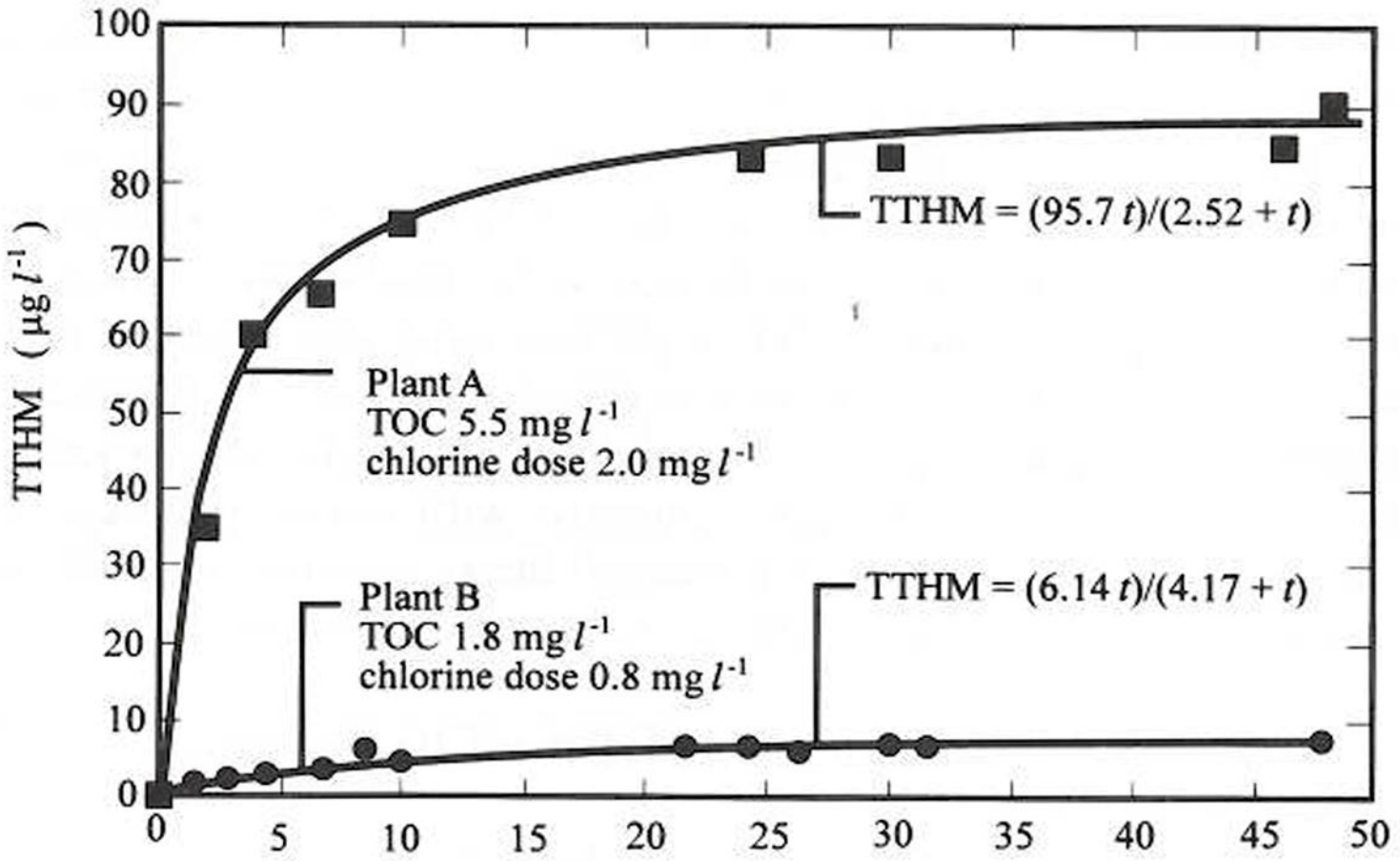
IMPORTANCE OF CONTACT TIME AND CHLORINE DOSE IN THM FORMATION

- The vast majority of TTHMs are formed following primary disinfection in the first 8 hours of contact with chlorine, usually in the main storage reservoir
- Chlorine doses and consequently THM formation are typically more significant in primary disinfection due to
  - the higher chlorine doses required to verify disinfection efficacy
  - where chlorine demand remains in incompletely treated water due to excessive remaining TOC in the water
- Following secondary chlorine disinfection, THM formation reactions become disinfectant-limited when the free chlorine residual typically drops below a free chlorine residual of 0.3mg/L
- Where organic matter has not been adequately removed or organic sediments exist in reservoirs and pipelines, booster chlorination facilities can result in increased THM formation

RELATIONSHIP BETWEEN THMs, TOC, CHLORINE DOSE AND CONTACT TIME

Extract from Casey TJ and Chua HK (1995) Trihalomethanes in Drinking Water - IEI Dublin



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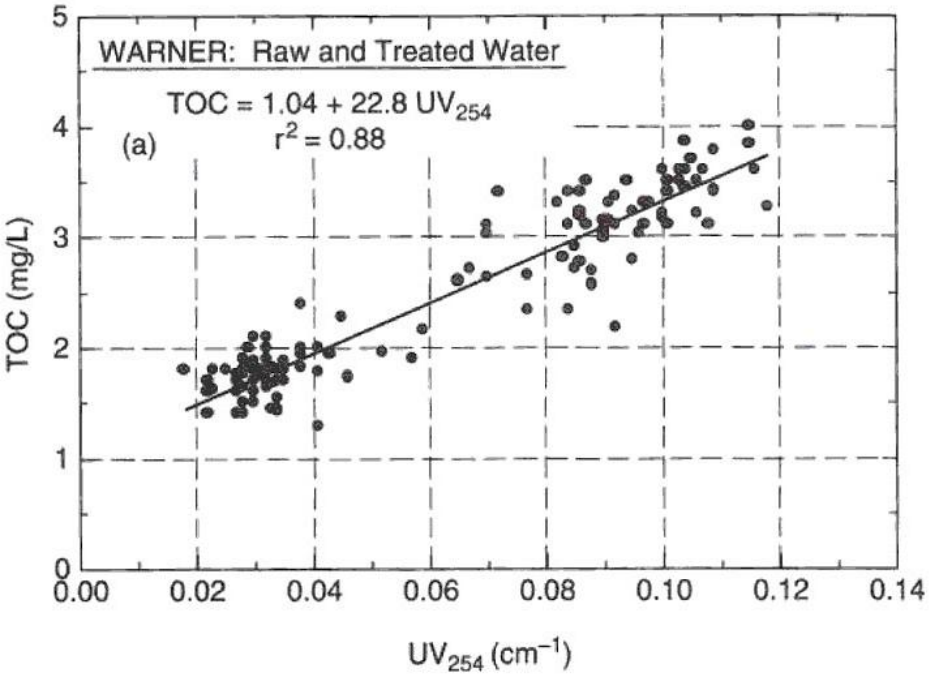
ASSESSMENT OF NOM IN WATER

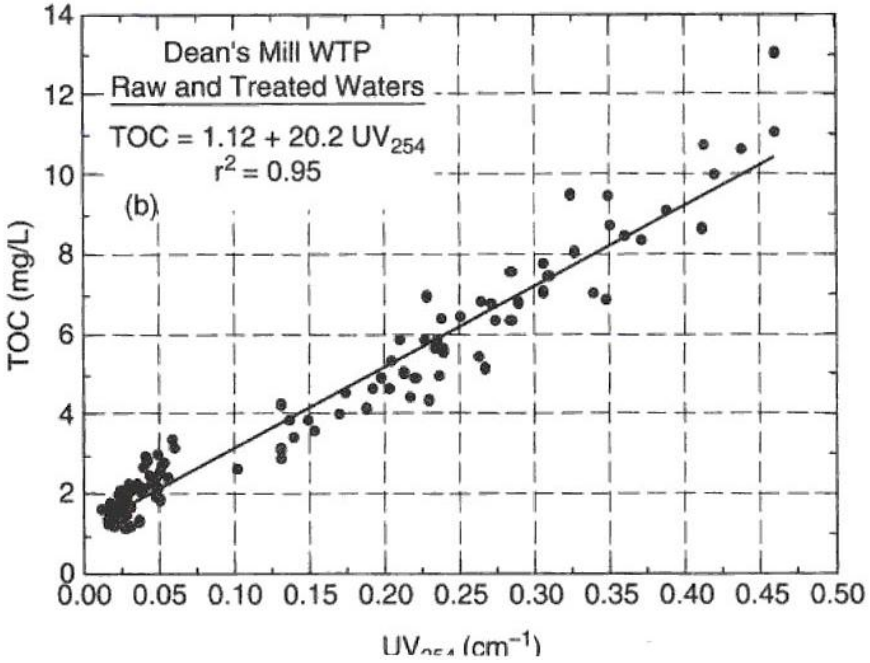
- Surrogate parameters which facilitate the assessment of NOM & the monitoring of water treatment plant operation and performance are :
  - Total and dissolved organic carbon (TOC and DOC);
  - UV absorbance at 254 nm wavelength (UVA_{254})
  - UV Transmittance (UVT) which is deducible from UVA_{254} in accordance with

$$UVA = 2 - Log_{10}UVT$$

- UV_{254} is related to the aromatic chain components of NOM and is considered a good predictor of humic TOC levels in water and the tendency of NOM in the water following treatment to form THMs after chlorination
- DOC usually constitutes > 90% of remaining TOC levels in filtered waters

UVA AS A TOC SURROGATE IN WATER





2009 ï Edzwald and Kaminski
Based on raw and treated water sampling results taken between 2002 and 2006

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SUVA AS AN INDICATOR OF NOM TYPE AND THM FORMATION POTENTIAL

- **SUVA is defined as the ratio between ultra violet absorbance (UVA) at 254nm (UV_{254}) & the DOC in the water**

$$SUVA = \frac{UVA_{254}\ (cm^{-1})}{DOC\ (mg/L)} \times 100 \quad (L/mg.min)$$

SUVA>4 indicates the presence of humic matter of high aromatic and hydrophobic character with high UVA (or low UVT), high chlorine demand and high THM formation potential (THMFP)

2<SUVA<4 indicates a mixture of humic and non-humic matter of both hydrophobic and hydrophilic character, medium UVA, lower chlorine demand and less THMFP

SUVA< 2 indicates a high fraction of non-humic matter of hydrophilic character with low UVA, a low chlorine demand and low THMFP

- **Optimal operation of coagulant based treatment systems for THM removal**

WHERE RAW WATER SUVA > 2.0, JAR TESTING AND COAGULATION DOSING IS BEST DETERMINED AND CONTROLLED USING UVA_{254} (as a surrogate for TOC) AND NOT TURBIDITY

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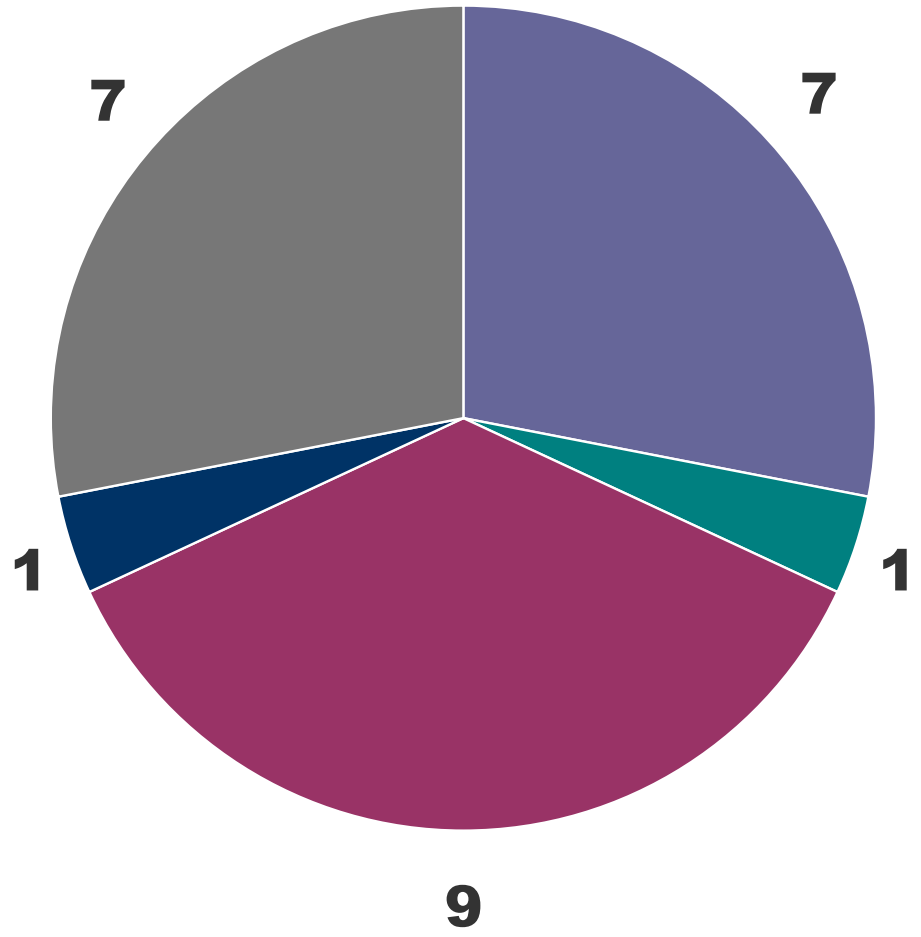
SUMMARY OF TREATMENT PROCESSES IN EPA PROJECT



- No Pre-Treatment
- Direct Filtration
- Slow Sand Filtration
- Ozone + GAC
- Conventional Treatment

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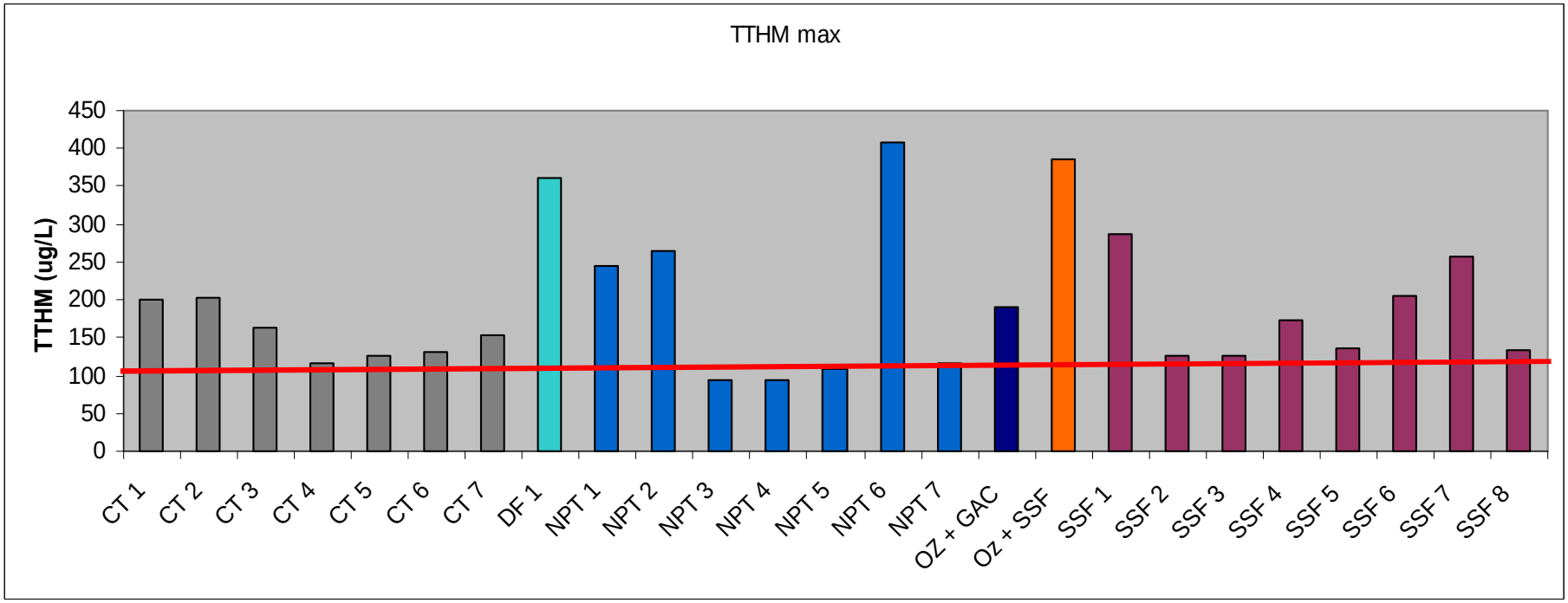
SUMMARY OF MAX TTHM LEVELS



No Pre-Treatment

Sand Filtration

Slow Sand Filtration

Ozone + Slow Sand Filtration

Ozone + GAC

Conventional Treatment

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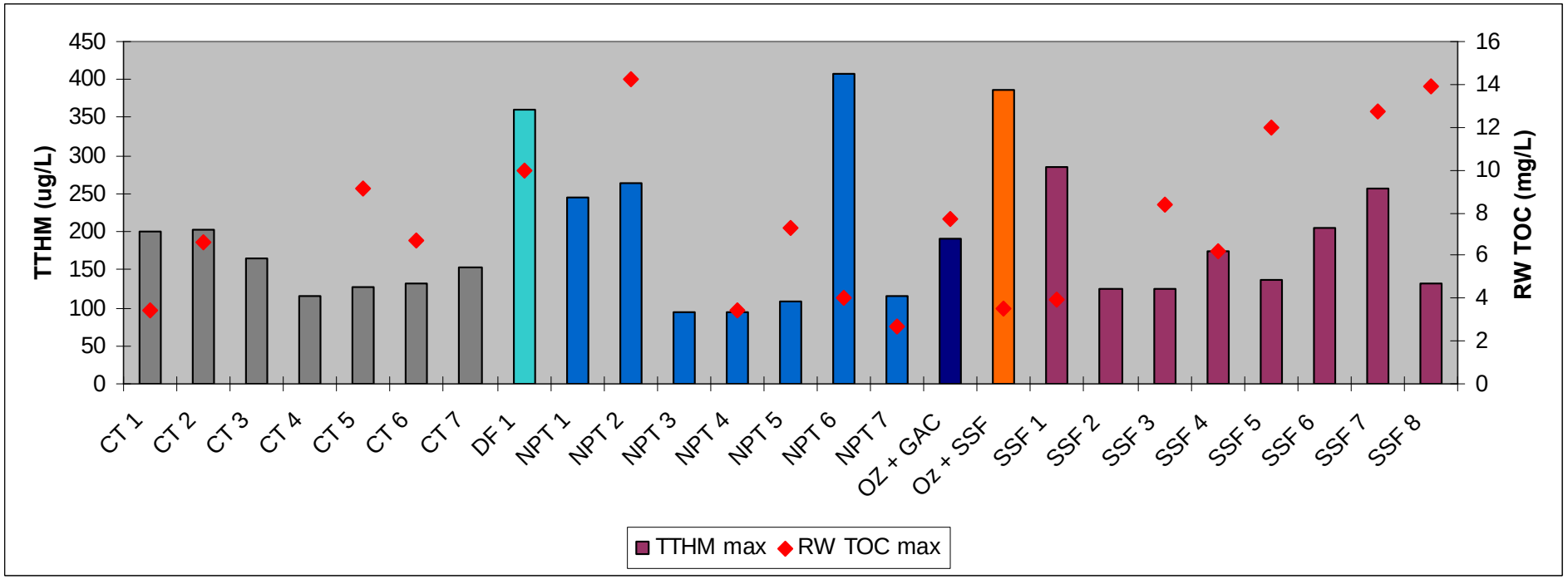
SUMMARY OF TTHM vs RW TOC



No Pre-Treatment

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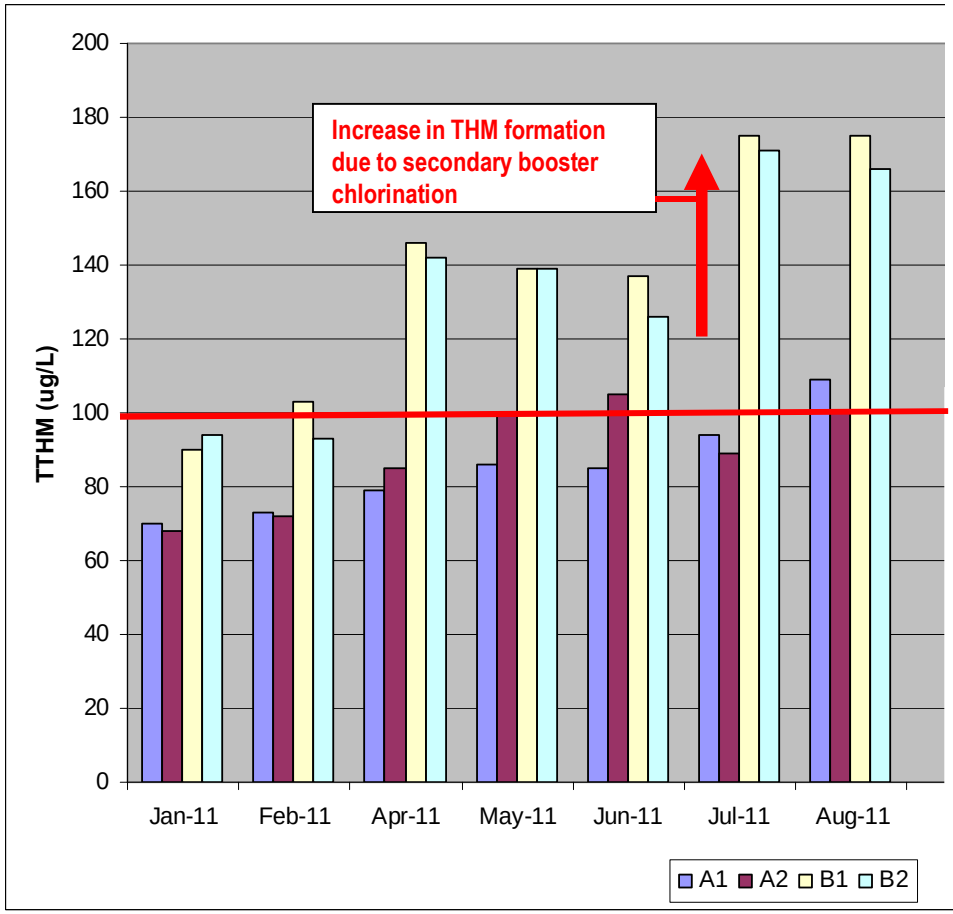
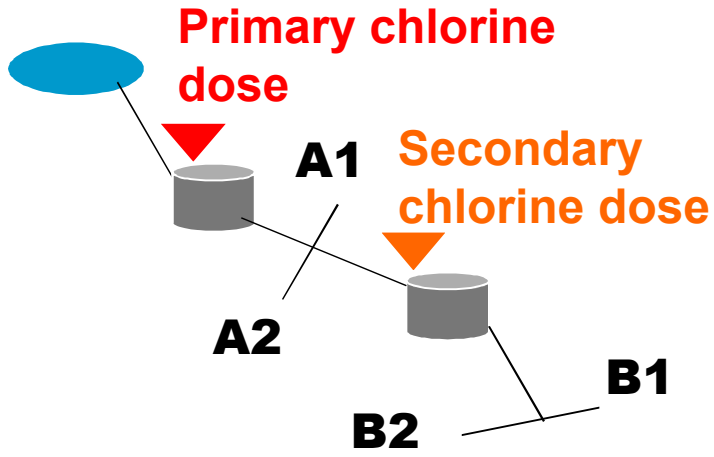
THM FORMATION INCREASE AFTER BOOSTER CHLORINATION

Case Study – SFF1



- Without effective treatment for NOM removal, THM formation in excess of parametric limits will continue to ensue following primary, secondary and booster chlorination.



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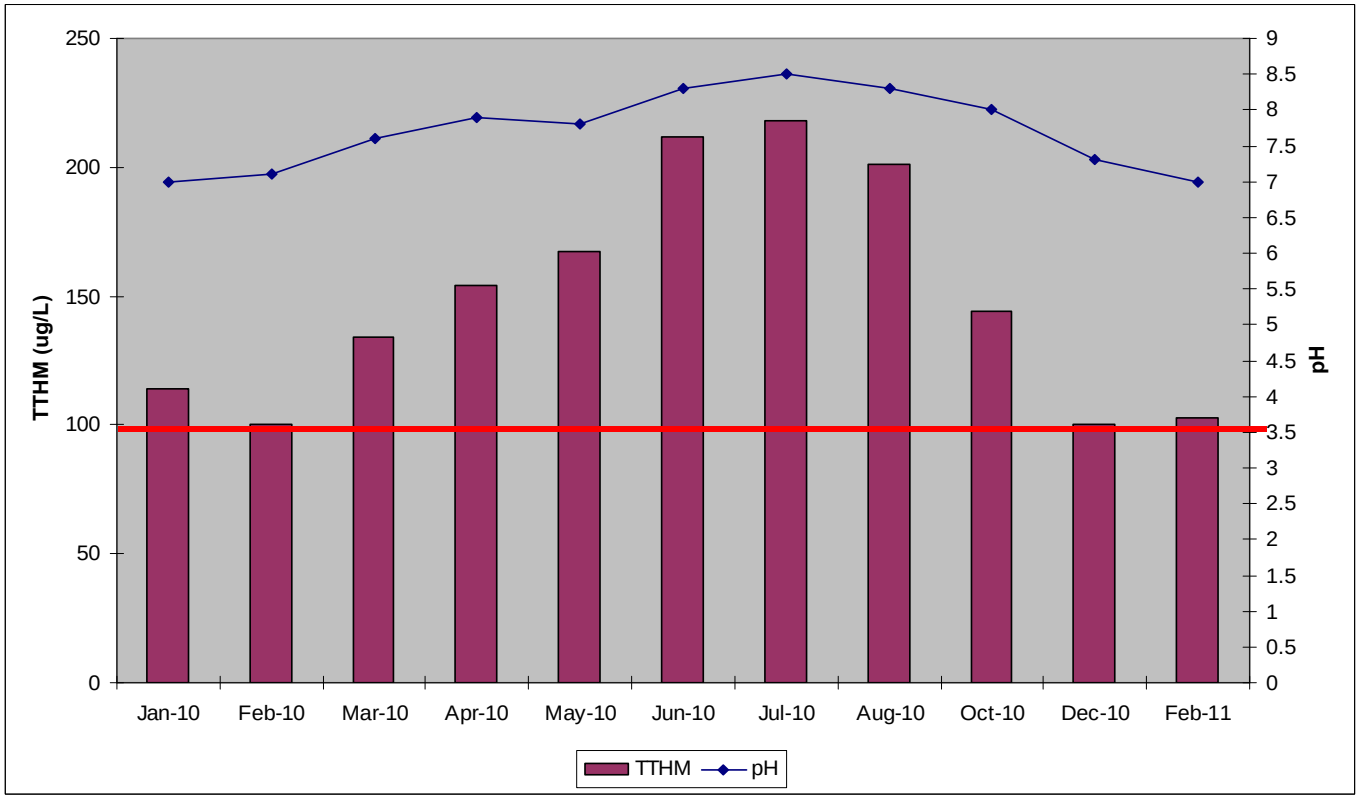
THM FORMATION INCREASES WITH RISE IN pH

Case Study – SFF1



Natural pH variation in water causing THM peak. pH adjustment should be provided to optimise chlorine dose at a pH ≤ 7.5.

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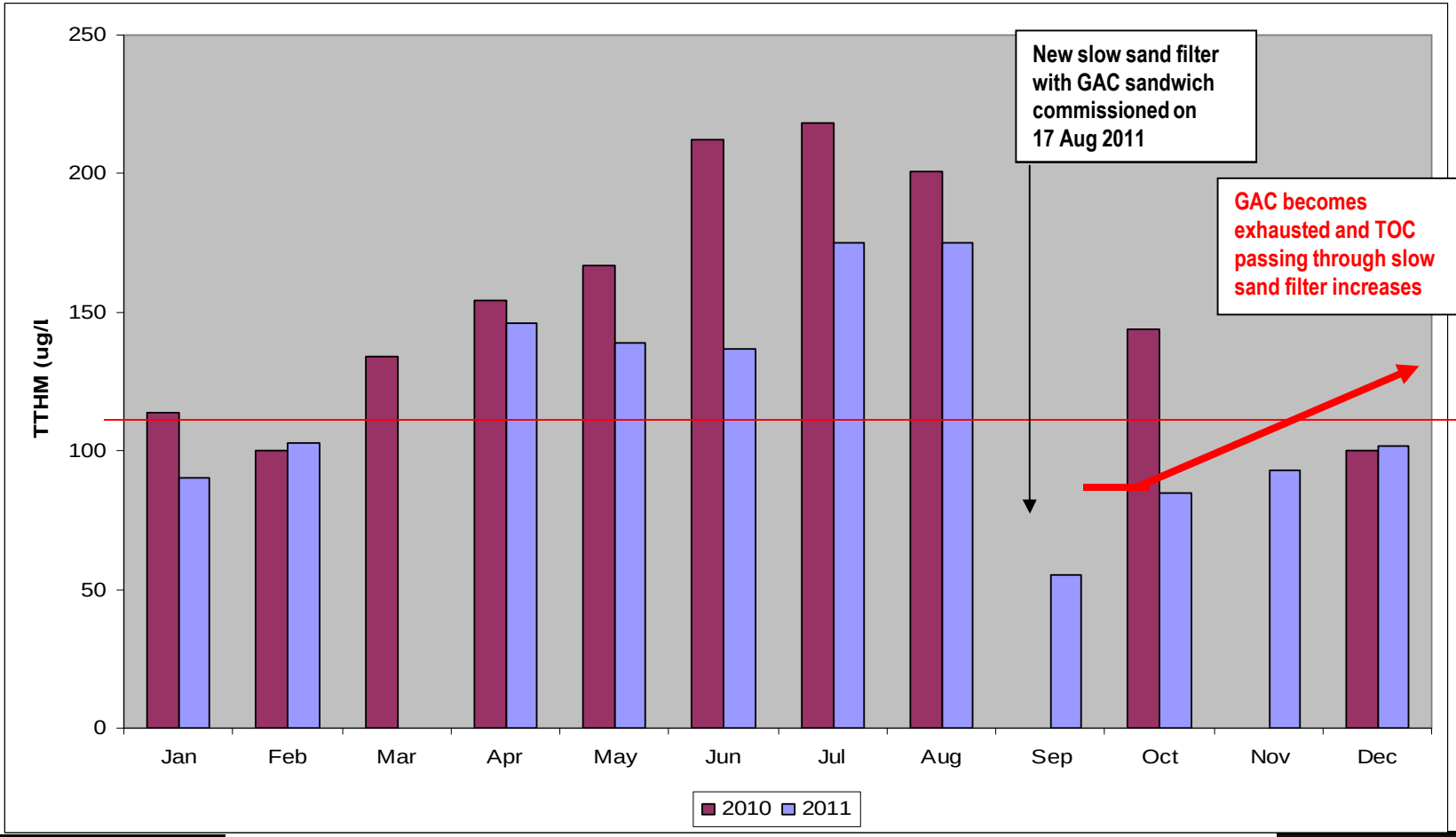
EPA NOM Conference

Meyrick Hotel, Galway

16th June 2016

SLOW SAND GAC SANDWICH – ADSORPTION vs BIOLOGICAL

Case Study – SFF4



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SOURCE PROTECTION

Case Study – NPT 5 (No pre-treatment)

- **2004 to 2008 – Spring source under the direct influence of surface water.**
- **2009 – WSA completed source protection works to exclude all surface water ingress.**

	TTHM	TOC
2004 to 2007	<5 - 109	4.5 ï 7.3
2009 to 2010	<6	<1

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OZONE + GAC BIOLOGICAL FILTRATION vs CONVENTIONAL TREATMENT

Treatment	Ozone and GAC (GAC operating as a biological filter)		Coagulation, sedimentation and filtration	
	RW	TW	RW	TW
TOC (mg/l)	7.7	7.1	7.4	3.8
DOC (mg/l)	7.7	6.3	7.3	3.0
UVA	0.47	0.26	0.46	0.09
SUVA (L/mg.min)	6.0	4.1	6.3	2.9
TOC removal efficiency		8%		49%
TTHM (mg/L) (reservoir outlet)		86		69

- Water to both plants was abstracted from same source producing water with colour less than 20 Hazen (Regular THM exceedances at ozone plant)
- Final water colour less than 20 Hazen is not an indication of the DOC fraction of TOC containing most of the THM precursors.

  (NB. Oxidation for colour removal does not remove organic contaminants)

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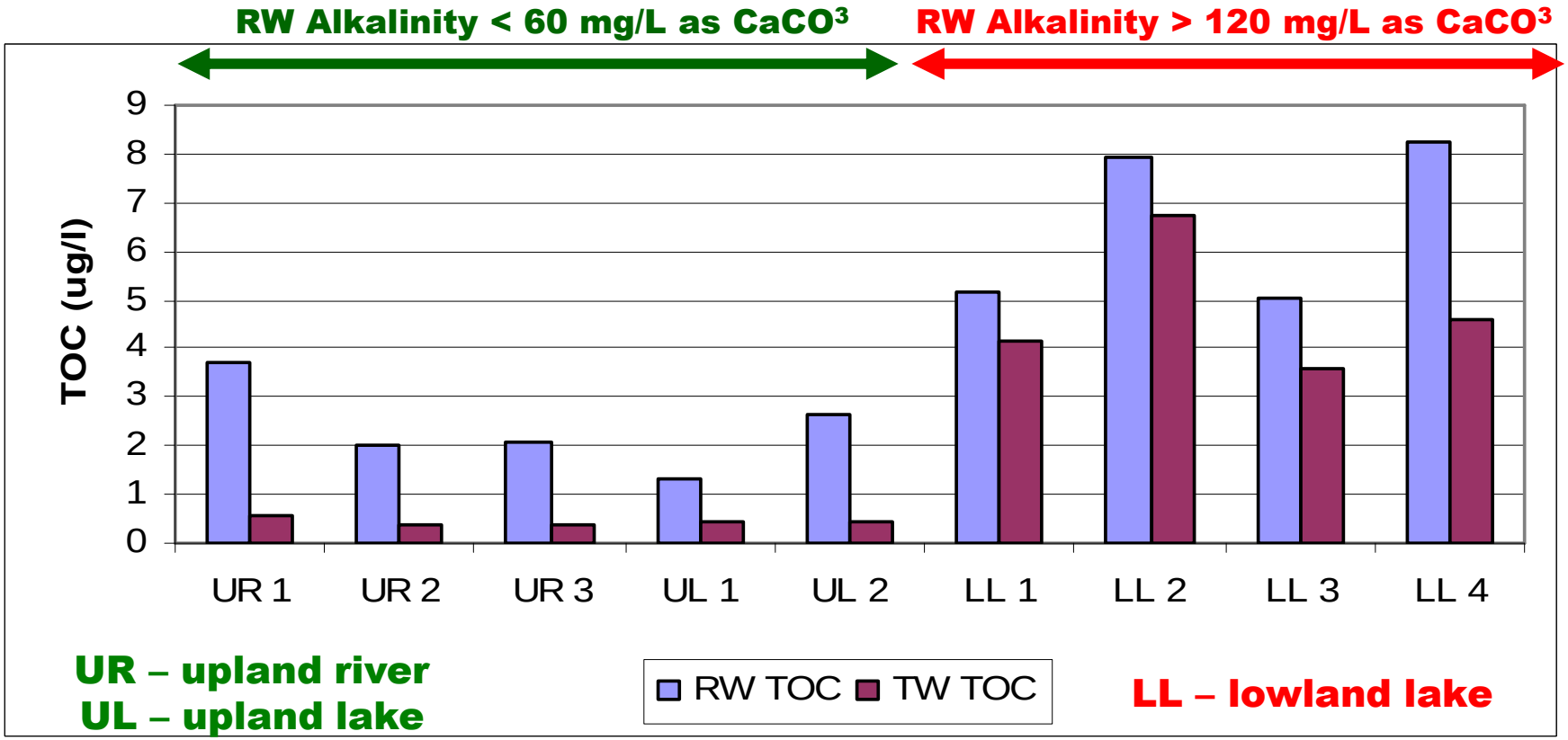
TOC REMOVAL EFFICIENCY – ULTRAFILTRATION MEMBRANES



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THE COLLECTION OF SITE SPECIFIC DATA ON THM FORMATION POTENTIAL

- **Where TTHMs levels in drinking water exceed the parametric value of 100µg/l, WSA should undertake**
  - Evaluate the effects of critical individual parameters on THM formation, their interaction, correlation and variability
  - Evaluation of the water supply system with regard to the treatment and distribution operational practices
    - **Stage 1 – Efficacy of THM precursor removal by treatment**
      - Stages 1a – Analysis of collated raw water and final TW sampling results to determine the TOC removal efficiency of the treatment process
      - Stage 1b – Determination of the nature and reactivity of THM precursors in the treated water using SUVA of the water from sampling and testing of its constituent water quality parameters namely UVA and DOC
    - **Stage 2 - Storage**
      - Operational goal for TW which should not exceed 0.08mg/l (80µg/l) TTHM
    - **Stage 3 – Distribution system**
      - Operational goal for TW which should not exceed 0.1mg/l (100µg/l) TTHM

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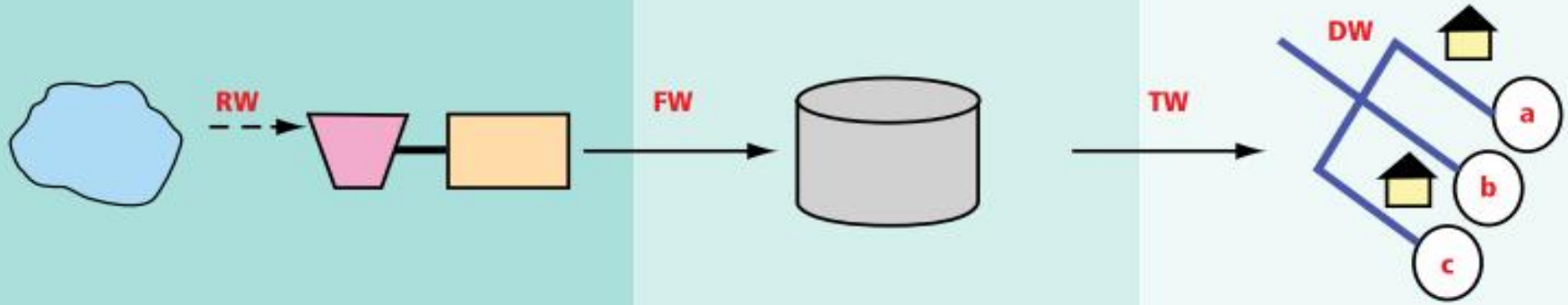
Source	Treatment Process	Storage	Distribution System



Stage 1a

Parameters to be monitored (Monthly)

1) FW TOC (mg/L)

2) RW TOC (mg/L)

3) RW Alkalinity (mg/L as $CaCO_3$)

Stage 1b

Parameters to be monitored (Monthly)

4) RW DOC (mg/L)

5) RW UV254 (/cm)

6) FW DOC (mg/L)

7) FW UV254 (/cm)

Stage 2

Parameters to be monitored (Monthly)

8) TW TTHM (mg/L)

9) FW total chlorine (mg/l)

10) TW chlorite (mg/l)

11) TW Bromate (mg/l)

12) TW free chlorine residual (mg/L)

13) FW pH

14) TW Temperature (0C)

15) Storage water age (hrs)

Stage 3

Parameters to be monitored (Locations within the network)

16) DW TTHM (mg/L)

17) DW free chlorine residual (mg/L)

18) Monitoring location water age

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POSSIBLE MITIGATION MEASURES TO CONTROL THM FORMATION

- Control over Cl_2 dose and contact time in storage reservoirs can be used to reduce THM formation consequent to Cl_2 primary disinfection dose rates required by IW for bacteria and virus inactivation
- As secondary Cl_2 boosting disinfection dosage does not require contact time, potential exists to reduce THM formation by application of dosage into storage outlet pipes rather than prior to reservoirs
- pH control to a level at or below 7.5 (if possible) increases disinfection efficacy resulting in less dosage. Elevated pH also increases THM formation
- The alternative use of ClO_2 is not feasible due to excessive formation of chlorites and chlorates at the high doses of ClO_2 required in waters with residual NOM
- Chloramination as an non-THM forming secondary disinfectant has potential for use in smaller schemes not interconnected with other supply schemes

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SUMMARY OF CONCLUSIONS

- The predominance of surface water sources means NOM removal is critical
- Highest levels of TOC exists in lowland lakes and rivers (variable)
- Possible health risks from THMs not solely a drinking water issue
- The key THM precursors in treated drinking water are humic acids which are in the non-visible DOC range of particle size (<0.45um)
- The management of THM exceedances requires an understanding of the factors causing THM formation following treatment/disinfection, the seasonality of THM precursor presence in sources and the THM formation potential during subsequent water storage and distribution
- UVA is a good surrogate for TOC levels in water and in particular the larger aromatic chain DOC THM precursor compounds (i.e. humic acids)
- The use of the SUVA parameter (derived from DOC and UVA) is an indicator of THM formation potential in raw water. Consequently UVA is better than turbidity and colour (in the POC range) as the target parameter for contaminant removal in lakes
- Oxidation systems do not remove organics – they only break up POC organics into a smaller biodegradable DOC - more reactive with Cl_2

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SUMMARY OF CONCLUSIONS

- Filtration systems (RGF or SSF) on their own prior to chlorination are totally ineffective for removal of DOC THM precursors which are smaller in size than the filter barrier they present (0.45µm and 0.25µm respectively)
- The use of GAC for removal of oxidised organics is only effective when used in the adsorption phase and is only cost effective when treating waters with TOC levels less than approx 2.5mg/l
- The proposed use by IW of UV as multi-barrier primary disinfectant with chlorination means that verification of UV reactors (as per Clause 12 of SI 122 2014) is also dependant on adequate NOM removal so as to exceed the minimum UVT level of the UV reactor validation certification
- Membranes treating high alkalinity waters achieve reduced THM precursor removal
- Coagulant based systems, optimally operated, are best at removing the humic THM precursors - optimal operation in high TOC surface waters (lakes) is best achieved by the use of UVA as the target contaminant in jar testing to determine coagulant dosing rates and also for process control
- THM mitigation measures should be evaluated on a site specific basis based on the calculation of THM formation potential of the water following treatment

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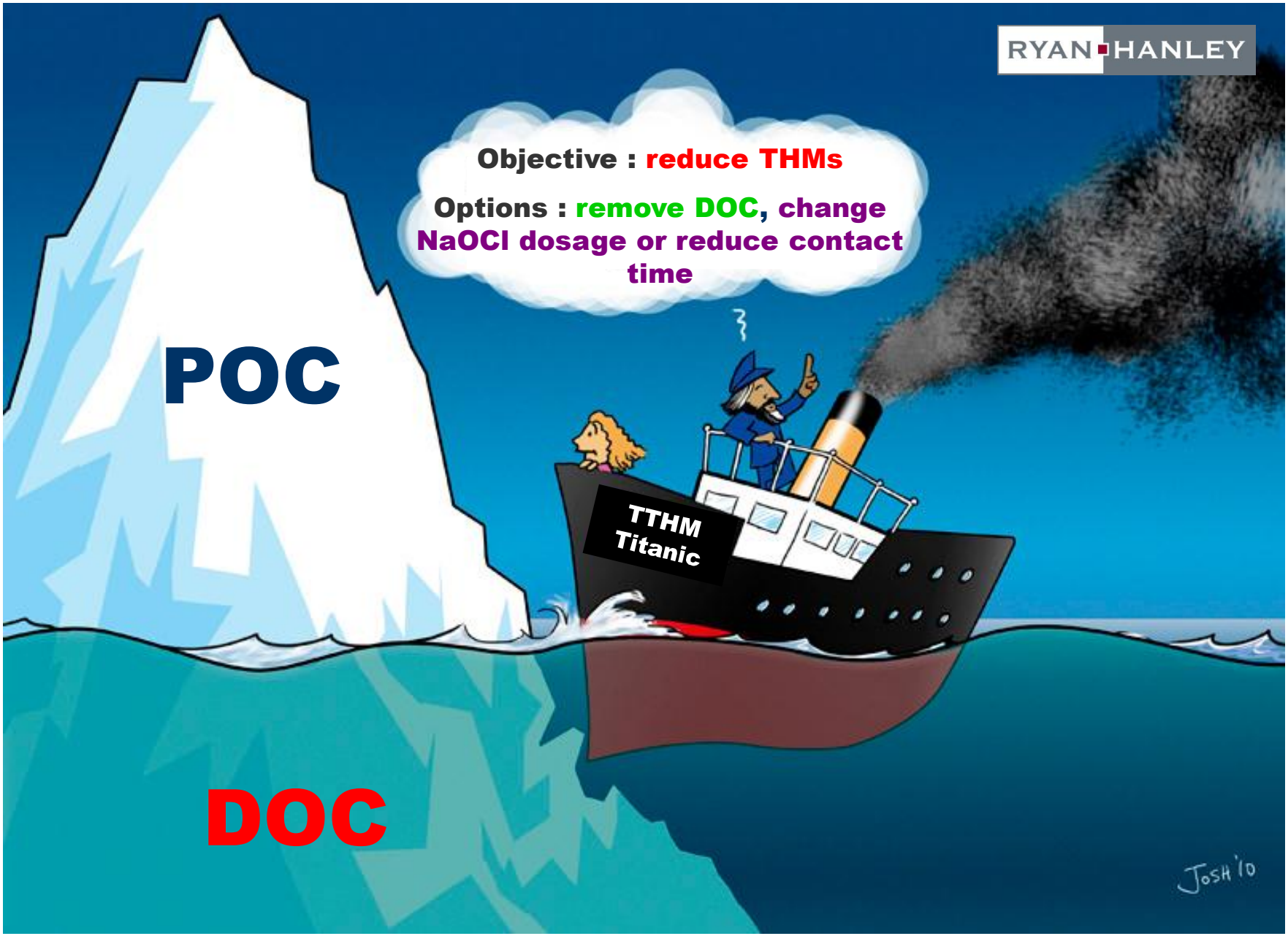
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Objective : reduce THMs
Options : remove DOC, change NaOCl dosage or reduce contact time
POC
DOC
TTHM
Titanic
Josh '10