# Disinfectant demand prediction using surrogate parameters – a tool to improve disinfection control

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## ABSTRACT

This paper describes the findings of a study to develop a "Disinfectant Demand Sensor". The aim was to determine the 3- or 7-day demand of a water sample in a shorter period of time, such as a few hours. Three approaches were taken to achieve this: (i) assessment of water quality surrogate parameters, (ii) "short" disinfection demand assessment, and (iii) temperature elevation approaches. All these methods can be used for the prediction of chlorine, but only temperature elevation and short demand assessment can be used for the prediction of chloramine with good accuracy.

Water quality data (prior to disinfection) was used to identify surrogate water quality parameters for disinfectant demand. The study found that the surrogate parameters of major interest were $UV_{254}$, DOC, and colour with $R^2$ values of 0.94, 0.91 and 0.83, respectively for chlorine demand prediction.

A rapid fractionation technique using ion-exchange resins to separate the organic component into various fractions demonstrated that the VHA (Very Hydrophobic Acid/humic acid) fraction showed the best relationship with chlorine demand and was also identified as a possible surrogate parameter with an $R^2$ value of 0.88. Chloramine demand did not behave similarly. Other factors influencing chloramine decay make it more difficult to predict than chlorine decay.

The possibility of using the demand of a shorter time span to predict demands of a longer time was also investigated. This technique was proved to work well for both chlorine and chloramine and results have shown that a 3-h chlorine demand correlated well with the 3-d demand ($R^2 = 0.97$). Also, the 1-d chloramine demand correlates with the 3-d demand ($R^2 = 0.92$).

Thirdly, the temperature investigation used the concept of raising the temperature of a sample thereby reducing the time taken to reach the 3-d demand equivalent. This method was found to be suitable for both chlorine and chloramine demand prediction. For example the 3-d chlorine/chloramine demand at 20°C equivalents of Myponga water can be determined after just 5.3/5.5 h respectively when the sample is incubated at 40°C.

**Key words** | chloramine, chlorine, demand prediction, disinfection, water quality

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## INTRODUCTION

The goal of most water quality managers is to maintain a safe water supply that is palatable to customers. Maintaining a disinfectant residual throughout the distribution system is an effective way of ensuring the aesthetic and bacteriological quality of the water at the customer tap. Chlorine is one of the most widely used disinfectants in drinking water treatment. The benefits of chlorine disinfection are well known and include low cost, a broad range of effectiveness and it has the added advantage of remaining active within the system for a considerable time. However, because of its ability to produce odours, the concentrations of chlorine and particularly the variability of

these concentrations is one of the most frequent causes of customer complaints. The conditions in Australia make control of disinfectant residual in distribution systems difficult. Problems arise from high levels of natural organic matter (NOM) that result in a high disinfectant demand, seasonal variation in flow demands and varying water temperatures. In addition, distribution systems are designed primarily for supply and fire fighting rather than optimised for water quality and hence long residence times may be found in water storage tanks and distribution pipelines. Monitoring of disinfectant residual in the water distribution system often suffers from delayed feedback which results in reduced responsiveness in disinfectant dose changes at the treatment plant. Consequently, applied doses that are either too high or too low are often identified too late for an operator to react and to take corrective action (Hua *et al.* 1999; Rodriguez & Serodes 1999).

Downstream of the treatment plant, chlorine gradually dissipates in the distribution system due to the reactions between various organic and inorganic compounds. The consumption of residual chlorine in the distribution system is influenced by: (1) bulk water reactions with organic and inorganic chemicals; (2) reactions with biofilms attached to the distribution pipe wall; (3) reactions between chlorine and corrosion deposits; and (4) mass transport of chlorine and other reactants between the bulk flow and pipe wall (Jadas-Hecart *et al.* 1992; AWWARF 1996; Vasconcelos *et al.* 1997). Generally these can be considered as chlorine decay in the bulk water and due to surface reaction (Kiene *et al.* 1998; Kastl *et al.* 1999*b*). Bulk chlorine decay rates due to chemical reactions in the aqueous phase can further be separated into fast and slow reactions. Fast reactions take place with easily oxidisable compounds such as inorganic compounds and are usually completed during primary disinfection at the treatment plant. Slow reactions proceed with less oxidisable compounds such as NOM (Dotson & Heltz 1985; Jadas-Hecart *et al.* 1992; Kastl *et al.* 1999*a*; Powell *et al.* 2000) and may occur in the distribution system. The bulk chlorine decay rates have also been observed to increase with temperature (Jadas-Hecart *et al.* 1992).

Conventional bulk water disinfectant demand methods are contact time dependent and often require several days to complete in order to match actual water ages found in water distribution systems. Knowledge of the bulk water

chlorine or chloramine demand is useful for a number of reasons. It can be used to assess water quality and to assist managers in optimising primary as well as secondary disinfection. It must also be determined in order to provide calibration constants if hydraulic models are to be used to predict disinfectant residuals in distribution systems.

In some cases, water quality prior to disinfection can vary markedly. Changes may be gradual, in the case of seasonal variation, or rapid if water is supplied from a number of sources. Unstable water supplies, having variable water quality, may exert a variable disinfectant demand. This requires substantial changes to be made to the applied disinfectant dose if disinfection residuals are to be maintained within the required ranges throughout the disinfection system. This presents a challenge to operators when attempting to control secondary disinfection as feedback loops are usually of the order of several days, making the process highly reactive.

This paper reports the suitability of a number of water quality parameters which were investigated as potential surrogates for disinfectant demand prediction. These will be utilised for the development of a rapid, on-line and generic "Disinfectant Demand Sensor" which can be used for a range of water qualities. The approach of this research was to formulate a relationship between the measurable surrogate parameters and disinfectant demand measured using the conventional methods over a set of water samples with a wide range of water quality. The main body of this study involved sample collection once every three months over a one year period to obtain waters from around Australia. The water quality including organic characterisation and disinfection decays of these samples were determined. Relationships between the disinfectant demand and these water quality parameters were established in order to identify surrogate parameters for both chlorine and chloramine.

Apart from using the measurable water quality parameters to predict long term demand (e.g. 3- or 7-d), two other approaches, using the disinfection demand of a shorter time (3-h) and elevating the temperature of a sample to above ambient level, were evaluated. The temperature elevation study was conducted to determine the feasibility of raising the temperature of a disinfected sample to decrease the time taken to reach the 3-d demand equivalent at ambient temperature. These approaches

provided three potential techniques of disinfection demand prediction.

## EXPERIMENTAL

### Sampling locations

Water samples from different locations in Australia, including New South Wales (NSW), Northern Territory (NT), Queensland (QLD), South Australia (SA), Victoria (VIC) and Western Australia (WA), were selected for this study. The water collection program was designed to collect a batch of 16 samples, comprising both raw and treated waters, from the selected locations every three months between 2003 and 2004. This selection produced a database of varying water qualities throughout Australia and incorporated seasonal variation. The database facilitated the determination of surrogate parameters for the prediction of disinfection demand that would hold over a range of water qualities and seasons.

### Analytical methods

General water quality parameters, pH (D-21 and 9620-10D, Horiba, Japan), turbidity (2100AN, Hach, USA) and dissolved organic carbon (DOC) (820, Sievers Instruments Inc., USA) and UV absorbance at 254 nm ($UV_{254}$) (Model 918, GBC Scientific Equipment Ltd., Australia) were determined using the methods described in *Standard Methods* (APHA 1998). Colour was determined using the spectrophotometric method described in Bennett & Drikas (1993). Ultrapure water used in these experiments was obtained from a Milli-Q® purification system (Millipore, France).

The rapid organic fractionation technique reported in Chow *et al.* (2004*a*) was modified just for the determination of the very hydrophobic acids (VHA) fraction (adsorbed by DAX-8).

Chlorine decay was determined by dosing an appropriate volume of saturated chlorine solution into the samples with pH adjusted to 7.2. For chloramination (monochloramine), ammonia then chlorine was added at a ratio of $4.5:1 = Cl_2:NH_3$ at pH 8.2 while mixing. Sample size was 2 litres and was stored in an amber bottle at $20°C \pm 2°C$. At predetermined times 100 mL samples were taken for chlorine/chloramine analysis over a period of 7 d. Chlorine residual was determined using the N,N-diethyl-*p*-phenylenediamine (DPD) titration method. DPD is used as an indicator in the titration procedure with ferrous ammonium sulfate (FAS) (APHA 1998). Chloramine residual was determined after the addition of a few drops of 5% potassium iodide solution (APHA 1998). For other decay temperatures (temperature evaluation study), a water bath with variable temperature control (Reciprocating Shaking Water Bath Model RW 1812, Paton Science Pty Ltd, Australia) was used.

## RESULTS AND DISCUSSION

### Sample selection

The water samples investigated in this study provided a wide range of water quality characteristics found in Australia and covered a DOC concentration range of 1.5–13.5 mg/L. The selection of the 16 sample batches was based on an earlier screening test (Chow *et al.* 2004*b*), which contained over 35 water samples. A summary of general water quality parameters, DOC, $UV_{254}$ and colour are presented in Table 1.

Treatment of some supplies consisted simply of disinfection while others used several clarification processes including coagulation/flocculation–sedimentation–filtration prior to disinfection. For those waters requiring treatment prior to disinfection (coagulation–sedimentation–filtration), both the raw and treated waters were included, whereas for water sources without the need of a clarification process, only raw water was used. The reason for using raw water was to provide a larger variation in water quality. All waters used for this study were collected before disinfection.

### Demand prediction using water quality parameters

NOM is usually represented by the measurement of total (TOC) or DOC concentration. The impact of NOM on various treatment processes is based upon both

**Table 1** | The range of water quality in this sample set used for this study

|  | Raw water | | Treated water* | |
| --- | --- | --- | --- | --- |
|  | Minimum | Maximum | Minimum | Maximum |
| DOC (mg/L) | 1.6 | 13.5 | 1.5 | 5.8 |
| $UV_{254\,nm}$ (cm$^{-1}$) | 0.035 | 0.632 | 0.022 | 0.151 |
| Colour (HU) | 4 | 127 | 2 | 13 |

*Treated water in this table included samples from water supplies with only disinfection is required (no clarification is needed).

concentration and character (Owen *et al.* 1995). Characterisation techniques, such as fractionation using resins and structural analysis using analytical instrumentation, have been developed worldwide to study the character of NOM. In particular, $UV_{254}$ is one of the simplest methods and is widely adopted by the water industry. It has been reported as a surrogate parameter to monitor the concentration of NOM (Edzwald *et al.* 1985; Wang & Hsieh 2001). In addition, $UV_{254}$ tends to give a measure of unsaturated organic bonds, which are potential sites with which chlorine can react. The UV absorbance of NOM in a water sample is potentially related to its chlorine demand (Powell *et al.* 2000). It can also provide a measure of overall disinfection by-product formation after chlorination (Korshin *et al.* 1997; Li *et al.* 1998).

## Single parameter prediction

Three general water quality parameters, DOC, $UV_{254}$ and colour, were examined individually and correlated well with the 3-d chlorine demand; DOC ($R^2 = 0.91$), $UV_{254}$ ($R^2 = 0.94$) and colour ($R^2 = 0.83$), as shown in Figure 1(a). The results have confirmed that a relationship exists between chlorine demand and the parameters DOC, $UV_{254}$ and colour. These parameters are potential surrogates for chlorine demand and, by using the regression equation of the graph, the chlorine demand may be determined by inserting the surrogate value into the equation of the line.

Figure 1(b) shows the predicted versus actual 3-d chlorine demand using the corresponding water quality parameters. These graphs have been established using data from a 12 month period and using a wide range of samples, so in theory these surrogate parameter graphs could be applied nationally. In addition, these findings hold over a wide range of samples and seasonal variations and are in keeping with Edzwald *et al.* (1985) and Chang *et al.* (1998).

NOM is a well known factor that can impact on chlorination and chloramination. The use of simple water quality parameters such as DOC, $UV_{254}$ and colour can provide some information about the organics. The application of an organic characterisation technique is a useful tool to obtain more detailed information on the NOM. However, most of the organic characterisation techniques are generally time consuming and rely heavily on very sophisticated instrumentation, which makes these techniques unsuitable for rapid assessment. However, a rapid fractionation method based on the selective adsorption of organic compounds with different chemical properties onto specific types of ion-exchange resin has been reported (Chow *et al.* 2004*a*). A further simplified version which determines just the VHA fraction was used to assess the predictability of disinfectant demand. The VHA fraction of organic matter, which is sometimes attributed to the humic acid fraction, showed the strongest correlation to the 3-d chlorine demand and so can be used in its prediction.

A complete set of regression analyses is presented in Table 2 with $R^2$ values of the correlation between water quality parameters against 1-h, 3-h, 6-h, 1-d, 3-d and 7-d chlorine demand. The results indicate that the water quality parameters correlate strongly with each step of the chlorine decay curve with the highest correlation being achieved for $UV_{254}$ ($R^2 = 0.94$), DOC ($R^2 = 0.91$), VHA ($R^2 = 0.88$) and colour ($R^2 = 0.83$) with the 3-d demand prediction.

These $R^2$ values show that it is not necessary to measure all three parameters to correlate water quality with chlorine demand. This is logical, given that $UV_{254}$, DOC and colour are generally understood to be interrelated.

In Figure 1(b), the plots of predicted versus actual chlorine demand using these parameters indicate a linear relationship and confirm that a reasonable prediction can be established. However, this general equation (including both raw and treated waters) may not be able to provide the required accuracy for water quality managers to predict chlorine demand. Statistical analysis by using $UV_{254}$ of just
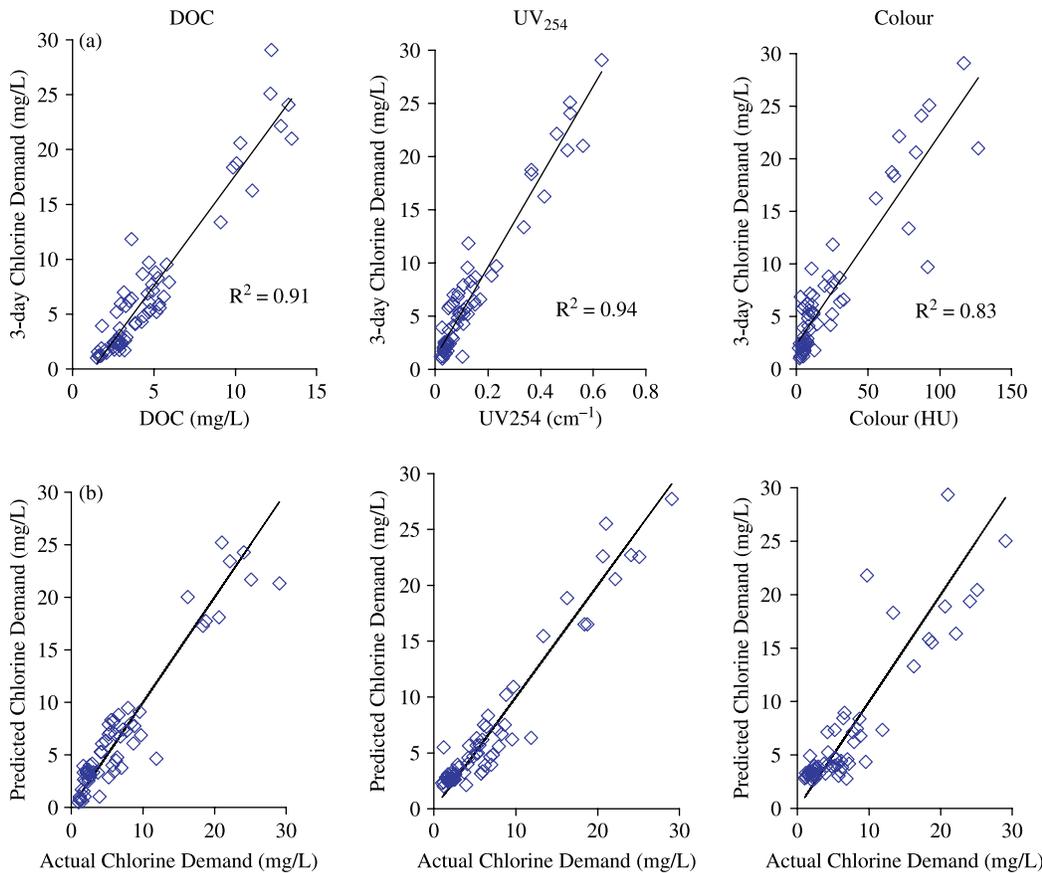
**Figure 1** │ Water quality surrogate parameters. (a) The relationship between 3-d chlorine demand against DOC, $UV_{254}$ and colour. (b) The predicted 3-d chlorine demand versus the actual 3-d chlorine demand using the corresponding water quality parameters.

**Table 2** │ The $R^2$ values of the correlation study of each water quality parameter against chlorine demand

|  | $R^2$ | | | |
| --- | --- | --- | --- | --- |
| **Chlorine demand** | **DOC** | **$UV_{254}$** | **Colour** | **VHA** |
| 1-h | 0.86 | 0.88 | 0.77 | 0.85 |
| 3-h | 0.87 | 0.90 | 0.79 | 0.85 |
| 6-h | 0.83 | 0.90 | 0.81 | 0.85 |
| 1-d | 0.89 | 0.93 | 0.83 | 0.87 |
| 3-d | 0.91 | 0.94 | 0.83 | 0.88 |
| 7-d | 0.90 | 0.94 | 0.85 | 0.87 |

the treated waters has confirmed that this fine-tuned equation has a prediction accuracy of better than $\pm 1$ mg/L chlorine demand (Figure 2). The data used to establish this relationship were analysed and outliers removed. The outliers were found to be consistently from two particular sources of water. One of the sources had ammonia present in the treated water prior to chlorination.

The presence of ammonia can impact on chlorine demand prediction as it reacts with chlorine to form chloramine. In order to get reliable chlorine demand prediction from $UV_{254}$, DOC or colour, it is worth checking the water is ammonia-free.

Similar experiments were conducted for chloramination. Chloramine demand did not correlate well with the measured parameters; for example $UV_{254}$ ($R^2 = 0.42$), colour ($R^2 = 0.39$) and DOC ($R^2 = 0.48$) for the 3-d
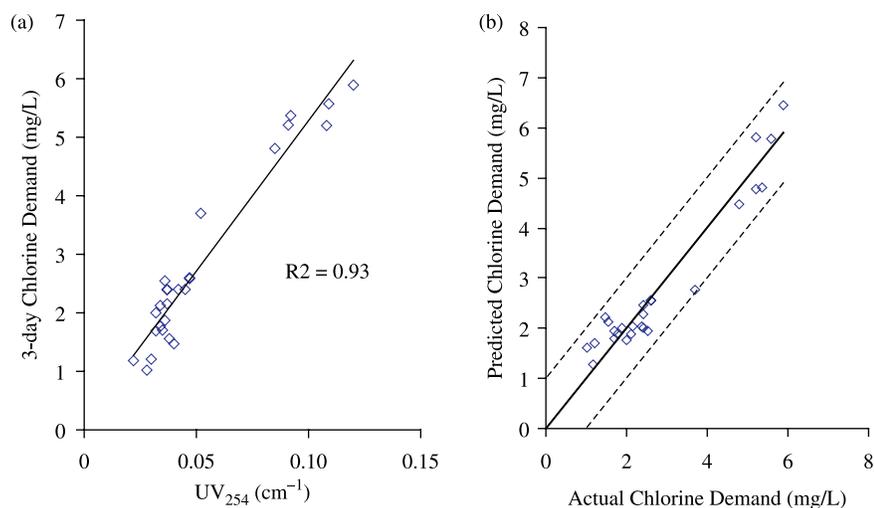
**Figure 2** | (a) Refined relationship between UV absorbance at 254 nm and 3-d chlorine demand. (b) The predicted 3-d chlorine demand using UV absorbance at 254 nm versus the actual 3-d chlorine demand, The dotted lines represent ± 1 mg/L chlorine demand boundary.

demand prediction (Table 3). Unlike chlorine demand correlation (Table 2), it appears that the chloramine correlation ($R^2$) is in the order of VHA > DOC > colour > $UV_{254}$ (Table 3). The mechanism of chloramine decay is certainly more complex compared with chlorine decay (Leung & Valentine 1994a,b; Vikesland et al. 2001; Duirka et al. 2005). A recent publication by Duirka et al. (2005) has confirmed there are two reaction pathways, monochloramine autodecomposition and reaction with NOM, which is rather complicated for monochloramine decay in the presence of NOM. Further work needs to be focused on

selecting alternative analytical techniques to identify the parameters related to chloramine decay.

## Multiple parameters

Instead of confining the prediction of disinfection demand to just a single surrogate parameter, a combination of the surrogate parameters can be used to predict chlorine. A multi-linear regression analysis using Excel's Data Analysis Toolbox (Microsoft, USA) allowed different combinations of surrogate parameters to predict chlorine demand.

**Table 3** | The $R^2$ values of the correlation study of each water quality parameter against chloramine demand

| Chloramine demand | DOC | $UV_{254}$ | Colour | VHA |
|---|---|---|---|---|
| | $R^2$ | | | |
| 1-h | 0.20 | 0.18 | 0.23 | 0.22 |
| 3-h | 0.23 | 0.20 | 0.26 | 0.26 |
| 6-h | 0.25 | 0.22 | 0.29 | 0.29 |
| 1-d | 0.35 | 0.31 | 0.35 | 0.40 |
| 3-d | 0.42 | 0.36 | 0.34 | 0.45 |
| 7-d | 0.48 | 0.42 | 0.39 | 0.53 |

**Table 4** | Multiple regression analysis for chlorine demand prediction

| Parameter combination | 3-d demand | 7-d demand |
|---|---|---|
| | $R^2$ | |
| $UV_{254}$, DOC, colour, VHA | 0.95 | 0.94 |
| $UV_{254}$, DOC, colour | 0.95 | 0.94 |
| Colour, $UV_{254}$ | 0.94 | 0.94 |
| $UV_{254}$, DOC | 0.95 | 0.94 |
| DOC, colour | 0.93 | 0.93 |
| $UV_{254}$ | 0.94 | 0.94 |

From the correlation results shown in Table 4, marginal improvement based on the $R^2$ value can be obtained by using multiple parameters. However, it can be concluded that not all parameters contribute to an accurate prediction to the same degree as simply measuring $UV_{254}$ produces almost the same correlation as using all four parameters.

### Short demand assessment to predict longer decay

White (1992) has reported a mathematical equation to predict chlorine demand at the end of any contact time, providing the chlorine demands at 1 h and at 30 min are known. In this part of the study, the concept of using short demand values to predict longer demand values is extended to samples with a wide range of water quality. Disinfectant decay curves were established over a duration of 7 d in this study with demands taken at 1, 3, 6, 24, 72 and 168 h. It was investigated if the determination of a 1-, 3- or 6-h disinfection demand could lead to the prediction of longer demand times such as multiple days. A simple regression analysis was performed by plotting the shorter demand against longer demand. A graph of 3-h chlorine demand against 3-d chlorine demand has shown that the 3-h results correlated with the 3-d demand with an $R^2$ value of 0.97 (Figure 3(a)). This confirmed that it is feasible to use a shorter demand to predict the demand after several days.

A detailed regression analysis is presented in Table 5.

A similar analysis was performed on chloramine decay and the results showed some potential for predicting 3- and 7-d demand using a shorter contact time demand. The most promising prediction was found using 1-d demand to predict 3-d demand ($R^2 = 0.92$).

### Temperature elevation approach

Temperature is an important factor that influences chlorine and chloramine decay. In this study, an attempt was made to take advantage of the fact that at higher temperatures disinfectant will decay faster. Understanding of this can also be used to predict decay caused by diurnal temperature fluctuations. In many parts of Australia, temperature variation can be very large (temperature can reach 45°C in mid-afternoon) which can have a significance impact on disinfectant residual control.

The effect of increasing the sample temperature on the time taken to reach the 3-d disinfectant demand equivalent was studied. Water samples from South Australia were used for this investigation and both raw and treated water samples were taken. Decays were set up at 20, 30, 40, and 50°C which allowed a calibration curve to be established for that water as shown in Figure 4.

This study showed very promising results for both chlorine and chloramine, as indicated in the graphs. The results show
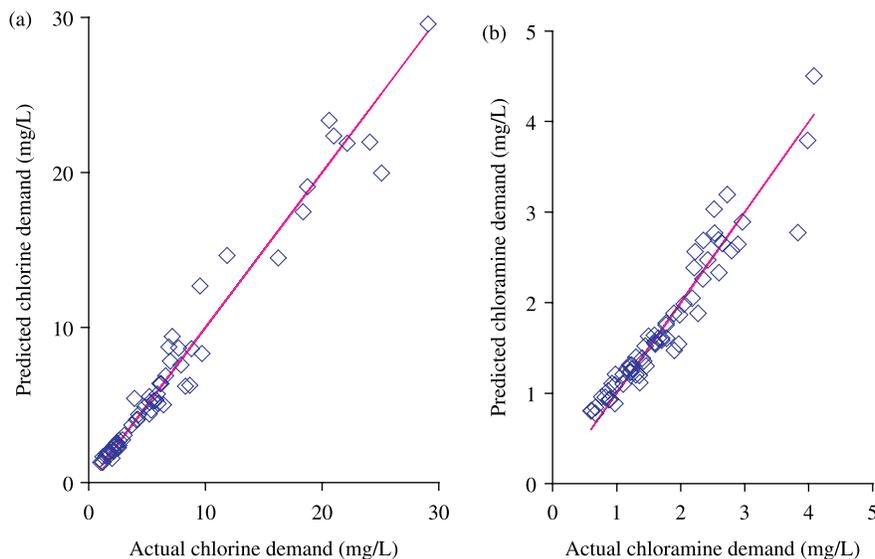


**Figure 3** | Predicting longer demands from shorter demand values. (a) Using 3-h chlorine demand to predict 3-d chlorine demand. (b) Using 1-d chloramine demand to predict 3-d chloramine demand.

**Table 5** | $R^2$ values for shorter demand time against longer demand times
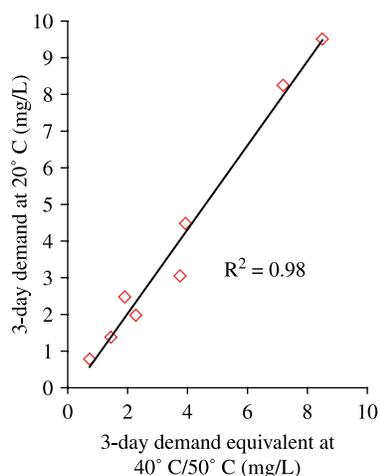
|  | 3-d chlorine demand | 7-d chlorine demand | 3-d chloramine demand | 7-d chloramine demand |
|---|---|---|---|---|
| 1-h demand | 0.94 | 0.91 | 0.69 | 0.60 |
| 3-h demand | 0.97 | 0.95 | 0.72 | 0.63 |
| 6-h demand | 0.97 | 0.95 | 0.77 | 0.68 |
| 1-d demand | 1.00 | 0.99 | 0.92 | 0.83 |
| 3-d demand | – | 0.99 | – | 0.90 |
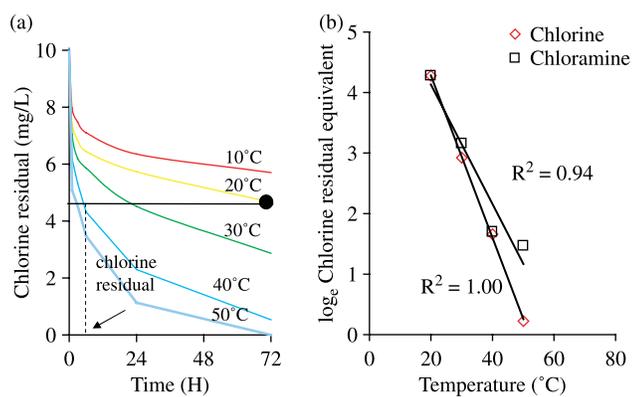


**Figure 4** | (a) Chlorine decay curves for Myponga (South Australia) water under different temperatures and an illustration of how to determine the chlorine residual equivalent. (b) Temperature calibration curves for chlorine and chloramine.



**Figure 5** | Validation of the actual 3-d disinfectant demand versus demand equivalent at 40/50°C.

that by elevating the temperature of a sample then dosing with disinfectant, a 3-d equivalent can be determined in a matter of hours, instead of waiting for three days.

For example, in the case of chlorine, Figure 4(a) shows the 3-d demand (ambient temperature 20°C) can be reached in just 5.25 h ($\log_e$ 1.65) when the sample is heated to 40°C. The same applies to the chloramine prediction for Myponga Water – Chloramine where the 3-d demand equivalent can be determine in just 5.5 h ($\log_e$ 1.7) when the sample is heated to 40°C.

Further validation work was carried out to test this approach for both chlorine and chloramine. Using the above calibration curves, the samples were heated and the demands were determined at the times indicated. Figure 5 shows a range of actual chlorine and chloramine demand results plotted against the values obtained at a higher temperature

and shorter reaction time. This illustrates the feasibility of this approach for both chlorine and chloramine.

It was established for this particular method of disinfectant prediction that an individual calibration is required for each water source. Testing for seasonal variation was not undertaken, but it may be assumed that the relationship is primarily temperature dependent and is not thought to be affected by any variations in organic matter. Further tests on a range of waters having differing chemical characteristics are required to confirm this test as a generic surrogate of disinfectant demand.

## CONCLUSIONS

Two commonly used disinfectants, chlorine and chloramine, have been studied with the aim of developing a tool to

predict their demand. It is clear from this work that it would not be feasible to apply the same demand "sensor" to both disinfectants. From the study, three methods for the prediction of chlorine and two methods for the prediction of chloramine have been identified.

The chlorine demand predictions using surrogate parameters (DOC, $UV_{254}$ and colour) and organic characterisation (the VHA fraction) have been based on a national database incorporating a large number of Australian water types and including seasonal variation. This therefore produces a chlorine prediction method which should be capable of being applied to any water in Australia at any time of the year. The use of the VHA fraction to predict chlorine demand is slightly more time consuming in that it cannot be measured online like $UV_{254}$. However, it may be superior for certain waters to produce a more accurate demand prediction. The use of multiple surrogate parameters in combination is another possible alternative to the use of just a single parameter. However, accuracy is not improved beyond $UV_{254}$ measurement as it also produced an $R^2$ value of 0.94 with the 3-d chlorine demand. From this study no surrogate parameter was identified which correlated well with chloramine demand over the range of water samples and seasonal variation. This probably explained why chloramine decay is strongly influenced by water chemistry (such as pH change) and the measured parameters selected in this work were not the main factors controlling chloramine decay. However, from this work, there does seem to be some link between the chloramine demand and the organic character (VHA) of the water. Further investigations may lead to the identification of a surrogate parameter for chloramine demand.

This study has also demonstrated that it is possible to determine the 3- and 7-d bulk water disinfection demand rapidly and accurately if the short term disinfectant demand (e.g. 3-h) is known. This method was found to work extremely well for chlorine demand. Rapid determination of chloramine demand (1-d) was also possible.

Finally, the elevated temperature method can be applied to both the chlorine and chloramine demand predictions. This method seems to be independent of water quality but requires an individual temperature calibration curve for each water source. Further investigations into this method could see the development of an automated system for use in the field.

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