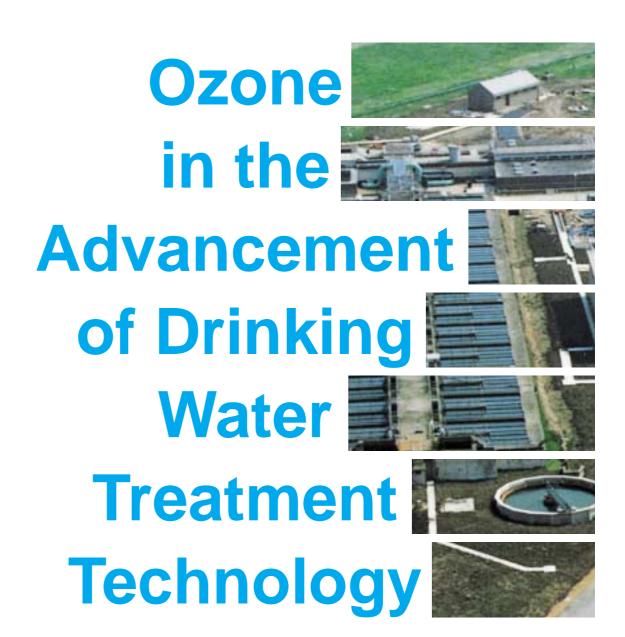
OZONIA



Ozonia – leading the way to a better world

Ozone in the Advancement of Drinking Water Treatment Technology

INTRODUCTION

In the course of the past two decades, drinking water treatment technology has advanced under the combined influence of three simultaneously occurring factors:

- a steady increase in the pollution of natural waters;
- a continued enhancement of the sensitivity and accuracy of analytical methods;
- a continual raising of drinking water standards (Figure 2 summarises some of the physicochemical criteria currently enforced by various regulatory bodies).

In the past, treatment lines differed greatly according to the characteristics of the water source involved. The aforementioned factors have further increased their diversity and complexity.



Figure 1: Coppermills Advanced Water Treatment Works Advanced ozone generators with a unit capacity of 48 kg/h at 10 wt%

Technical trends over the past few years have been particularly marked by:

Figure 2: Standards for key physicochemical characteristics of drinking water

	EU D	WHO	
Parameter	Guide Level	Max. Admissible Concentration	1993
Temperature (°C)	12	25	Acceptable
Turbidity (NTU)	0.4	4	5
Colour (Hazen units)	1	20	15
Odour (Threshold number)	0	12°C : 2	Acceptable
Taste (Threshold number)	0	25°C : 3	Acceptable
рН	6.5 - 8.5	-	6.5 - 8.5
Chlorides (mg·I-1 CI)	25	-	250
Sulphates (mg·l-1 SO ₄)	25	250	250
Aluminium (mg·I-1 Al)	0.05	0.2	0.2
Dry Solids at 180°C (mg·I-1)	-	1500	1000
Nitrates (mg·I-1 NO ₃)	25	50	50
Nitrites (mg·I-1 NO ₂)	-	0.1	3
Ammonium (mg·I ⁻¹ NH ₄ ⁺)	0.05	0.5	1.5
Permanganate value (mg·l-1 O ₂)	2	5	-
Hydrocarbons (mg·l-1)	-	0.01	0.001 - 1
Detergents (mg·I-1)	-	0.2	-
Iron (mg·l ⁻¹)	0.05	0.2	0.3
Manganese (mg·l-1)	0.02	0.05	0.1
Fluorine (mg·l-1)	-	0.7 - 1.5	1.5
Arsenic (mg·l-1)	-	0.05	0.01
Pesticides (µg·l-1)			
- per substance	-	0.1	0.03 - 100
- total	-	0.5	-

- the phasing out of chlorination in the main oxidation steps and the introduction of chlorine at the end of the treatment line for residual profection purposes;
- the more widespread use of ozone and the development of new ozone generators. Typically these operate on a medium frequency power source (500 to 1000 Hz) and may be oxygen fed as well as working at high to very high ozone concentrations. This trend has led to a gradual reduction in the size of equipment, a simplification of ancillaries, an increase in the generated ozone concentration and a decrease in capital and operating costs. Ozonia achieved a major breakthrough in early 1991 when it introduced to the market a new generation of ozone equipment, the "Advanced Technology" series. The enhanced performance of this family is attributed to the use of a new, non-glass dielectric material, which was the result of extensive research;
- the use of ozone at various stages of treatment.

CURRENT ISSUES IN WATER TREATMENT

There are certain problems in water treatment of either a chemical or a biological order that prove particularly difficult to correct. Some of these relate primarily to surface waters – although they can also affect groundwater.

Trace inorganic contaminants

One of these problems is the pollution from trace inorganic contaminants that are mainly discharged to the environment in industrial effluents [1, 2]. Some, such as Aq, As, Cr (III), Sn and Zn, can be effectively (90 to 100%) removed using conventional clarification processes. Others, such as Cd, Co, Cr(VI), Cu, Hg, Ni and Pb, require additional treatment including polishing by a combination of ozone and Granular Activated Carbon (GAC) or, in some cases, two consecutive clarification steps at different pH levels (Ni, Pb, Zn). It must be remembered that for most of these substances drinking water standards have set limits ranging from 1 µg·l-1 (Hg) to 50 μ g·l⁻¹ (As, Cr, Mn, Ni, Pb). There is a higher tolerance for iron, copper and zinc.

Dissolved aluminium is rarely found in excessive quantities in raw water. Higher aluminium concentrations are sometimes found in waters that have undergone treatment using an aluminium salt where the flocculation pH has not been accurately controlled (between 6 and 7.2). This problem can be avoided by accurately controlling the pH valve (i.e., the pH adjustment chemical must be added with the coagulant) or by replacing the aluminium sulphate with ferric chloride or polymers.

Trace organic contaminants

Most of these compounds are effectively removed by a complete treatment line beginning with preozonation and ending with the combination of ozone plus GAC. Special mention must be made of herbicides and pesticides, particularly atrazine and other triazines. Atrazine is:

a compound often present in surface waters and sometimes found in groundwater;

- not highly toxic (much less so than dieldrine, parathion, etc.) but is subject to the same limit values in drinking water as the other pesticides, i.e., 0.1 μg·l⁻¹.

In fact, atrazine is a pesticide that is extremely difficult to remove, as indicated by the following:

- 0 to 15 % in clarification (up to 20 % if preoxidation is applied);
- 15 to 40 % after ozonation, depending on the applied dosage and the contact time;
- 25 to 45 % in slow-sand filtration [3];
- 70 % after clarification with Powdered Activated Carbon (PAC) and ozonation [3];
- 90 to 100 % after filtration on GAC, but with a carbon service life of generally less than one year for the concentrations generally found in raw waters [4].

By contrast, the combination of ozone and hydrogen peroxide ($O_3 + H_2O_2$) produces hydroxyl radicals that results in the effective destruction of atrazine as well as the other organic compounds that resist destruction by ozone alone. All that is needed for this is a small dose of hydrogen peroxide (about 0.4 g per g of ozone). This technique alone achieves atrazine removal rates ranging from 70 to 85 % when used in conjunction with filtration on GAC to complete the treatment.

Trihalomethanes

This is a phenomenon involving trace level organics which are produced during treatment in what might be termed "induced contamination." During treatment chlorine reacts with certain functional groups of naturally occurring organic compounds (humic acids, algal metabolites, etc.), resulting in the production of volatile organohalogens that remain present in the treated water [5, 6]. The most common compounds are haloforms or trihalomethanes which have the general formula CHX₃ (where X = CI, Br, etc.).

Such as:

- chloroform (CHCl₃), which is also the most prevalent;
- bromodichloromethane (CHBrCl₂);
- dibromochloromethane (CHBr₂CI);
- bromoform (CHBr₃).

Other organohalogens are:

- carbon tetrachloride (CCl₄);
- trichloroethylene (C₂HCl₃);
- chlorobromomethane (CH₂BrCl);
- tetrachloroethylene (C₂Cl₄);
- dichloroethane (CH₂CI-CH₂CI).

There are two explanations for the presence of such bromide compounds. Bromine can be introduced as an impure component of industrial chlorine, or more often, by the oxidation of bromides present in raw water to HOBr, which is highly reactive in the presence of organic matter.

Definitions

Trihalomethanes are generally referred to under the abbreviation THMs, and their concentrations generally expressed in $\mu g \cdot l^{-1}$.

- Total trihalomethanes (TTHMs) refers to the sum of all THMs present in water at any given time;
- "Precursors" is a term which refers to the organic substances susceptible to reactions with chlorine to form organohalogens. These precursors are generally naturally occurring organic substances:
 - humic or fulvic acids;
 - intra or extracellular algal metabolites. The algal biomass is a significant source of THM formation.
- Trihalomethane formation potential (THMFP) refers to the maximum quantity of THMs that will be produced by the precursors present in the water, taking into account the conditions most favourable to THM formation (excessive free chlorine combined with a 3 or even a 5-day contact time). This is the most pessimistic hypothesis but reflects conditions which can actually occur in a supply system. Determination of this parameter is therefore of utmost importance.

THM Formation

The organics that react most readily with chlorine to form THMs are the polyhydroxybenzens, especially the metadihydroxylated derivatives.

Ketones can also lead to the same type of reaction, but with less favourable reaction kinetics. The phenomenon may be described by the basic reaction:

$$2R\text{-CO-CH}_3 + 6CIOH \rightarrow$$

 $2R\text{-CO-CCI}_3 + 6H_2O$

followed by the hydrolysis reaction:

$$R-CO-CCI_3 + H_2O \rightarrow$$

R-CO-OH + CHCI₃

Humic acids are primarily composed of chains of polyphenolic nuclei and alicyclic di-ketones. According to the above, these, in addition to algal metabolites, constitute the main THM precursors to be found in natural waters. Moreover, a positive correlation has been observed between the true colour of raw water and the quantity of THMs formed after chlorination.

The THM concentration in treated water depends on the chlorine dose applied and reaches maximum levels when the dose exceeds the breakpoint. THM concentration also depends on:

- pH (with maximum levels in alkaline media)
- contact time (increases gradually with contact time)
- temperature (increases with temperature)
- precursor content, of which the Total Organic Carbon (TOC) measurement can provide a clear indication.

Regulating THMs:

Minimising potential risks

The analytical techniques that now allow accurate measurement of THM levels did not become common until the early 1970s, which explains why the THM phenomenon was not detected until 1974. However, there is no doubt that the same phenomenon has occurred ever since drinking water chlorination began [7].

According to toxicological studies [8], the THM concentrations in water

treated with chlorine (in the order of $100 \, \mu g \cdot l^{-1}$):

- do not entail a risk of high toxicity;
- may entail a risk of long-term toxicity, particularly as a result of chloroform, due to its carcinogenic and mutagenic effects. However, the actual danger threshold seems to lie at levels above the concentrations usually encountered. In fact, statistical analysis has proven quite difficult in this area and has given rise to much controversy.

Therefore, at present, only potential risks may be considered, but they must not overshadow the much more tangible risks to public health, particularly those of a microbiological nature. It has been concluded that disinfection is of primary importance to water treatment, and that treatment lines must be modified to minimise the potential risks described.

This is the main issue of certain regulations with regard to THMs:

- in the United States, the Environmental Protection Agency has set a Maximum Contaminant Level (MCL) of 100 μg·l⁻¹ for TTHM in drinking water. The new by-products rule cut this down to 70 μg·l⁻¹ and will reduce this even further to 40 μg·l⁻¹ in the year 2002.
- in France, the decree enforcing the European directives on drinking water standards (January '89) does not set a mandatory limit. However, a public health policy was nonetheless defined in a circular (May 16, 1989) recommending the same maximum concentrations as those stipulated in 1984 by the World Health Organisation for a number of organohalogen compounds, particularly 30 μg·l¹ for chloroform (although the WHO relaxed their recommendations somewhat in 1993).

When comparing these mandatory or recommended limits, it must be taken into consideration that where chlorine is used in high doses, some municipal water supplies contained up to 300 $\mu g \cdot l^{-1}$ of CHCl₃, in addition to about 100 $\mu g \cdot l^{-1}$ of each of the other THM compounds (CHBrCl₂, CHBr₂Cl, CHBr₃). In France, where chlorine is applied as residual doses in the order

of 1/10th of those applied in the USA, THMs were reported at much lower concentrations, although levels still sometimes surpassed the limit values.

Another fact to bear in mind is that THMs are volatile compounds and represent a minor portion of all organohalogens that could form as a result of chlorination. The other portion consists of the non-volatile compounds, which are much more difficult to detect, even though they account for the main amount of the total organohalogen compounds.

As the potential risks associated with these other compounds are not currently known, THMs serve only as an indicator. Therefore, among other objectives, modern treatment techniques aim to prevent the formation of THMs, a goal that has led to significant changes in the design of drinking water treatment plants.

Combating THMs:

Removal or prevention?

THM molecules are relatively small in size and, consequently are difficult to remove by conventional processes. Clarification has practically no effect and ozone has only limited effectiveness. Only GAC is able to capture THM molecules, but its capacity to capture this particular type of molecule is rapidly exhausted. Full-scale experimental studies conducted on waters from the Loire and Seine showed the following flow ratings between the GAC regeneration cycles:

- 4 200 m³ of water per m³ of GAC, when the latter is used as a single filtration step, i.e. in the place of sand. For a rate of up to 7 volumes of water per hour and per volume of carbon, this corresponds to a regeneration interval of less than one month;
- 10 000 m³ per m³ of GAC when the latter is used as a second step following sand filtration and perhaps ozone, which corresponds to a service life of two months – still unacceptable in economic terms.

Air stripping offers a possible alternative, but would not remove non-volatile compounds and is a costly process that in most cases would also

raise complex problems related to the carbonate balance.

However, as chlorination has proven to be an essential component for water treatment, the approach taken has been preventive, rather than curative, based on two series of measures:

- elimination of prechlorination with postponement of all chlorination until the end of the treatment line. In this case compensatory measures are necessary, such as:
 - covering sedimentation and filtration tanks;
 - nitrifying the ammonium on filters;
 - or, breakpoint chlorination at the end of the treatment line.

An additional alternative is to use a preoxidation treatment that does not lead to THM formation, such as:

- chloramines, which are rarely used outside the USA;
- chlorine dioxide [7], which raises some doubts as to the innocuity of a number of by-products formed during treatment [9];
- or, preozonation, which offers many advantages including a reduction in THM precursors [10], which is therefore becoming the predominant preoxidation technique);
- optimisation of treatment processes with a view to maximising the removal of precursors before chlorination. This approach can be subdivided into two complementary strategies:
 - improvement of coagulation-flocculation-clarification-filtration lines;
 - polishing to remove residual dissolved organics from the treated water and, in turn, form as little THM as possible in the course of final chlorination prior to distribution. The treatment combination [O₃ + GAC] yields the best results.

Tastes and odours

Pollution can cause unpleasant tastes and odours in raw water. The main source of these problems actually lies in the metabolic activity of aquatic organisms [11]; the main source of these sapid and strong-smelling substances are Actinomycetes and blue-green algae (Cyanophyceae, also known as Cyanobacteria). The dominant metabolites are geosmine and 2-methylisoborneol, which produce earthy-musty odours. Many other substances are likely to be produced by different types of algae, which convey various types of odour to the water when they proliferate: grass, fish, must, pharmaceutical, violet, cucumber, etc..

In order to obtain drinking water that has no taste or odour, these substances can be:

- destroyed by oxidation. Ozone is generally the only oxidant that is effective for this application, although its action is limited because it sometimes has difficulty in attacking saturated compounds like geosmine or 2-methylisoborneol. Alternatively, a treatment combination of O₃ + H₂O₂ can completely destroy these compounds [12];
- or removed by adsorption on PAC or GAC. The latter is the most efficient solution, especially when used as a second filtration step, following ozonation of the water to extend the service life of the GAC [11].

Toxic metabolites produced by Cyanophyceae (or Cyanobacteria)

A number of blue-green algae (Microcystis aeruginosa, Anabaena flos-aquae, Aphanizomenon flos-aquae, etc.) have the disadvantage of producing toxic substances, which fall into two categories:

neurotoxins, which are alkaloids.
 Figure 3 shows the structure of one of these, anatoxin-a, which is similar to cocaine;

hepatotoxins, which are polypeptides.

Although these substances can cause unpleasant symptoms to people swimming in natural waters (dermatitis, conjunctivitis, etc.), there is little risk of their reaching a dangerous toxic level in a water supply. However, they might be at the root of specific symptoms of chronic toxicity (particularly due to the release of metabolites into the water once the algae have died off, which occurs when a body of eutrophic water has undergone treatment using copper sulphate):

- Hepatital symptoms in the form of increased levels of gamma-glutamyltranspeptidase in plasma [13] were reported in hospital patients in Armidale (Australia), where the water supply was contaminated by Microcystis aeruginosa blooms;
- Pyrogenic reactions were reported in patients of a blood dialysis centre coincident with a sharp increase in the Cyanophyceae content of the raw water [14].

These toxins passed through conventional clarification systems. However, they can be destroyed by ozone or removed by filtration on GAC. This constitutes a further reason for making increasingly systematic use of polishing treatments involving $[O_3 + GAC]$, particularly for waters with high algae contents.

Disinfection

Recent years have seen the reemergence of ancient diseases (like cholera in some tropical areas) and the discovery of new ones (such as gastro-enteritis caused by the protozoa *Giardia* and *Cryptospo-*

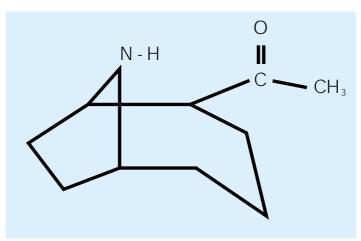


Figure 3: Anatoxin-a

Micro-organisms	Ozone pH: 6 to 7	Chlorine pH: 6 to 7	Chloramine opH: 8 to 9	Chlorine Dioxide pH: 6 to 7
E. Coli	0.02	0.03 - 0.05	95 - 180	0.4 - 180
Poliovirus 1	0.1 - 0.2	1.1 - 2.5	770 - 3500	0.2 - 6.7
Rotavirus	0.006 - 0.06	0.01 - 0.05	2810 - 6480	0.2 - 2.1
Giardia lamblia cysts	0.5 - 1.6	30 - 150	750 - 2200	10 - 36
Cryptosporidium	2.5 - 18.4	7200	7200 (1 log)	78 (1 log)
(oo)cysts				

Figure 4: C. T. factors necessary for a 99% (or log 2) inactivation at a temperature range of 5 - 25 °C (the lower the temperature the higher the C. T.)

ridium). Therefore, all water treatment specialists are in agreement that disinfection must be considered "top" priority in the preparation of drinking water.

Good physicochemical disinfection is based on the "C.T." factor, i.e. the product of the residual disinfectant concentration C (mg·I¹) by the contact time T (minutes). The C.T. value is thus expressed as mg·min·I¹ or g·min·m³. T should be considered as the true contact time in the contactor, determined (by means of a tracer) as the retention time for 10% of the water flow. At this T_{10} time, 90% of the water is still in the contactor but account is taken for the fraction which has been treated with the shortest contact time.

If T_h is the theoretical hydraulic contact time, calculated by the ratio volume to flow, the T_{10}/T_h ratio is very variable, ranging from less than 0.2 for a tank without baffles, up to 0.7 for a baffled tank.

The estimated values of C.T. necessary to obtain an inactivation of 99% (or log 2) of the main pathogenic organisms, are given in Figure 4. These clearly show that ozone is, by far, the most powerful disinfectant and particularly, the only one able to ensure an efficient inactivation of the cysts and oocysts of parasitic protozoa.

Optimising the application of ozone in drinking water treatment

It is, as far as disinfection is concerned, most probable that the future will see the selection of new indicators (in order to better assess disinfection efficiency), more widespread use of ozone and a better combination of disinfection with clarification.

In addition trends in micropollutant removal will focus primarily on advanced oxidation technologies, based especially on enhancing the effectiveness of ozone by combining ozonation with:

- another oxidant (especially H₂O₂)
- a catalyst (e.g., TiO₂, ZnO)
- a photocatalyst (e.g., UV, UV + TiO₂)

Again, the best results are obtained using the combination of ozone with hydrogen peroxide $[O_3 + H_2O_2]$. This combination triggers the release of free OH° radicals which are extremely active on trace organics as well as on unpleasant tastes and odours. However, there is a drawback with use of the $O_3 + H_2O_2$. It leaves no residual

ozone creating a problem when disinfection is also a requirement. To offset this, the use of the Degrémont patented three compartment contact design can be used, which:

- satisfies the chemical demand in the first compartment;
- disinfects the water in the second compartment, and
- performs radical oxidation of the trace contaminants in the third compartment, following the application of hydrogen peroxide in a proportion of 0.4 g to 1 g ozone.

Preozonation will also be increasingly applied. The merits of preozonation replacing prechlorination are:

- savings in coagulant dose;
- improvement of flocculation;
- better TOC removal;
- algae removal to the same level as with prechlorination;
- no THM formation;
- better reduction of THMFP;
- oxidation of Fe²⁺ and Mn²⁺, if present;
- partial removal of colour and odour;
- preparation of DOC and degradation of ammonia for the subsequent filtration.

Figure 5: Anglian Water's Grafham Water Treatment Works. Incorporating preozonation, final ozonation and Biflux GAC contactors.



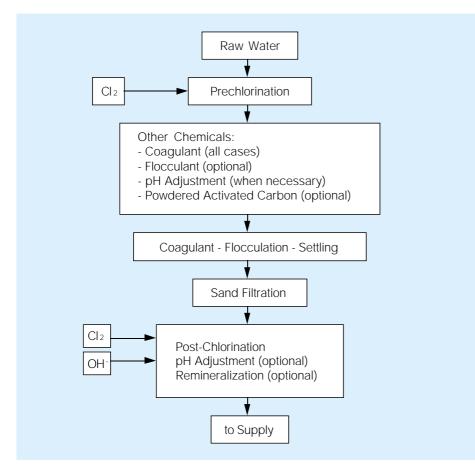


Figure 6: Conventional treatment line applied to surface waters in the 60s and 70s.

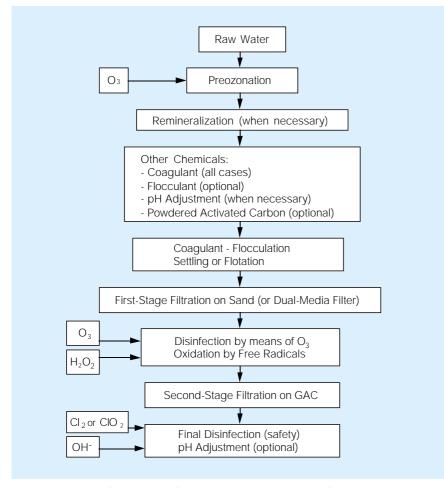


Figure 7: Typical treatment line for surface waters in the 90s

IMPACT ON THE DESIGN OF SUR-FACE WATER TREATMENT LINES

During the 60s and the early 70s, facilities that treated surface water (rivers, lakes, and reservoirs) were generally designed based on the diagram shown in Figure 6.

As shown, prechlorination had become systematic. Clarifiers and filters thus operated in a chlorinated environment which prevented virtually all biological activity. For example, the ammonium ion was removed by physicochemical process (breakpoint chlorination).

The preceding explains why the design of water treatment lines has evolved considerably over the past two decades, and why, considering the latest technical achievements, the most effective process for the treatment of surface waters that are polluted and/or contain high levels of THM precursors is felt to be the one that combines preozonation with a post-treatment polishing consisting of $O_3 + GAC$.

This treatment line is illustrated in Figure 7 and operates according to semi-physicochemical, semi-biological principles.

There is no doubt that whilst producing a better quality and safer drinking water, modern treatment lines are more complicated than the treatment typically applied during the 60s and 70s, described in Figure 6. However, progress in ozone generation over the last decade (medium frequency power supplies, high ozone concentration, oxygen feed), as well as the cost reductions in GAC, have changed the preconception that the cost must also be higher.

The costs of ozone plants were not only drastically reduced with the introduction of oxygen fed plants compared to the air fed plants of the 70s but also in the last years by introduction of revolutionary methods of ozone production. Figure 9 shows the increase of concentrations over the period 1990 to 1996 that could feasibly be produced at approximately the same energy consumption level. The result is an enormous saving in oxygen. A plant producing ozone at 6 wt% will need double the amount of oxygen than one producing

at 12 wt%. Today's water treatment plants work with ozone concentrations well over 10 wt% which represents a saving in oxygen of 40% in comparison to the usual of 6 wt% of the late eighties. Since this is at the same power consumption level and is combined with much lower maintenance costs then it is clear that the operating costs are drastically reduced.

Figure 10 illustrates the reduction of the energy consumption costs for a given ozone concentration during the same period showing the progress of the new technology.

At the same time the investment costs of the ozone generators for major plants dropped to fifty percent of the 1990 level (Figure 11) due to increased efficiencies of the new technologies.

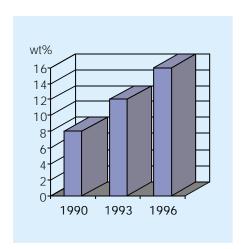


Figure 9: Progress of economic range of ozone production concentration

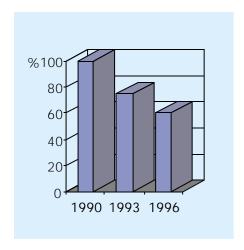


Figure 10: Progress of energy costs (basis 8 wt%)



Figure 8: Rostock Water Works. Advanced Technology ozone generators (3 x 8.7 kg/h at 10 wt%)

SAVINGS WITH OZONE

Resulting from the enhanced microflocculation effect of ozone the filtration rates can be increased, smaller filtration beds used and even the chemical consumption lowered. Because of the higher oxidation potential of ozone it is also possible to reduce contact times, unless of course it becomes desirable to enhance the biological effect on GAC. The fact that some substances can be attacked only by ozone favours the use of ozone even more. Cryptosporidium parvum for example, can be economically and effectively reduced in numbers with the help of ozone (Figure 4). All other economically applicable methods act only as barriers to the (oo)cysts but do not actually attack them.

As previously mentioned, ozone has not only a positive effect on COD removal, by breaking down refractory compounds and making them biodegradable, but ozone also prolongs the service life of the GAC. This alone makes ozone economically feasible when GAC is needed.

Ozone can totally replace chlorine, chloramine or chlorine dioxide in the preoxidation and mainoxidation stages. In addition, although some form of chlorine residual is nearly always required in the distribution networks, ozone can drastically reduce its use, enhance the quality of the water and still be more economical than other oxidants. Today new drinking water works treating contaminated water and designed according to the new concept, as described in Figure 7, are lower in investment as well as in the running costs. These saving have been demonstrated in many cities around the world, such as Los Angeles, Barcelona, Singapore, Paris and Zurich and are even more significant with the new ozone technologies.

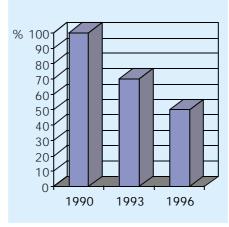


Figure 11: Progress of investment costs of major plants (basis 8 wt% production concentration)

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Figure 12: The control room of the Coppermills Advanced Water Treatment Works (GB) with the medium frequency power supply unit on the right



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