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Project Summary

Biological Processes in the Treatment of Municipal Water Supplies

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Studies were conducted of a European water treatment technique that appears to produce high quality drinking water without the synthesis of halogenated organic materials during the water treatment process. This biological treatment technique involves the sequential application of chemical oxidation (usually by means of ozone), rapid media filtration, optional reaeration, and granular activated carbon (GAC) adsorption.

The use of this biologically enhanced, granular activated carbon (BEGAC) technology was studied in several European water treatment plants to determine its advantages and disadvantages for use in the United States. Seven European water works were visited where chemical preoxidation is followed by rapid media filtration and GAC adsorption.

The process is still under development in these European water works, but results to date are positive. They indicate that in those water works using GAC adsorption of dissolved organic materials, incorporation of chemical preoxidation with small amounts of ozone (1 to 2 mg/L) can result in extending the life of GAC adsorbers by factors of 4 to 6 before reactivation is required. The process can be used for the biological removal of ammonia from raw water supplies and has replaced breakpoint chlorination in several European plants. Such replacement eliminates the prechlorination step, which in turn eliminates the formation of significant quantities of halogenated organics.

Results to date indicate that after biological equilibrium is attained in GAC adsorbers, 25 to 35 percent of the influent dissolved organic carbon is removed from solution biologically.

This Project Summary was developed by EPA's Municipal Environmental Research Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

This project resulted from the current need to learn as much as possible about methods for controlling organic contaminants in drinking water. The present project is an outgrowth of an earlier study on the state of the art of ozone and chlorine dioxide technologies in municipal water treatment (Public Technology, Inc. 1976. An Assessment of Ozone and Chlorine Dioxide Technologies for Treatment of Municipal Water Supplies. EPA-600/2-78-147. U.S. Environmental Protection Agency, Cincinnati, Ohio). During the course of this 4-week, onsite survey of European water treatment facilities, the site team observed the use of a biological treatment technique in France, the Federal Republic of Germany, and Switzerland that is not currently practiced in the United States. This technique involves the deliberate promotion of aerobic biological growths on filter media (sand, anthracite) and GAC media (columns or beds) for purposes of nitrification and of removing organic chemicals. The aerobic biological activity appears to be enhanced by an oxidation step applied before the activated carbon treatment. Such preoxidation steps frequently involve the addition of ozone.

Evidence obtained from numerous European pilot-plant studies and from several full-scale operating plants in Europe supported the claim that a properly designed and operated combination of ozone and GAC unit processes enhances the removal of some types of organic chemicals and can reduce the frequency of regeneration of the activated carbon media, depending on the reactivation criteria. The latter characteristic is vital, since one of the primary concerns of public water supply systems regarding the use of GAC (in addition to the high capital cost) is the relatively high cost associated with frequent reactivation. Furthermore, this biological process replaces breakpoint chlorination and eliminates the generation of chlorinated organic materials during the early stages of the water treatment processes.

Some experiences with the BEGAC process were described briefly in the earlier report (EPA-600/2-78-147), but additional details were required to establish this method as a viable means of enhancing treatment effectiveness and reducing operating costs. This study was therefore undertaken to acquire information on the following specific subject areas:

- Determining design criteria used for BEGAC systems in Europe;
- Determining mechanisms by which BEGAC systems operate;
- Determining microbiological aspects of BEGAC systems;
- Gathering field operational and cost data on BEGAC systems;
- Quantification of technical and cost benefits of BEGAC systems;
- Determining changes in U.S. treatment plant designs required for retrofitting BEGAC systems into existing plants.

Site Visits

After consulting with leading European water treatment authorities during early 1978, the site visit team conducted visits to selected European facilities during June 1978. The primary questions to be answered were:

1. Is BEGAC effective for removal of organic chemicals, and if so, under what conditions?
2. Is BEGAC an effective replacement process for ammonia removal by breakpoint chlorination, and if so, under what conditions?
3. How and why does the BEGAC process achieve its effectiveness?
4. Is a preoxidation step necessary? If so, must the preoxidant always be ozone?
5. Can the added capital and operating costs of an ozonation or other preoxidant system be offset by the increase in operating time before the GAC must be regenerated?
6. Is BEGAC bacteriologically safe to use for drinking water treatment?
7. What pretreatment and post-treatment steps are made necessary when BEGAC is incorporated into a drinking water treatment system?
8. Does biological regeneration of the GAC truly occur, and if so, to what extent?

Not all of these questions were answered, since the BEGAC process still is being studied and developed by European water treatment specialists. But answers to some of these questions were obtained by conducting a review of the published literature and a 3-week site visitation of the following facilities:

1. Operational drinking water treatment plants using granular activated carbon facilities designed to promote biological growth,
2. Research institutes and universities conducting studies on the BEGAC process, and
3. Activated carbon and ozone systems manufacturers in Western Europe during June 1978.

The scope of this report could not be confined to ozone/GAC treatment systems alone, however. Early in the study, it became apparent that Europeans use many biological processes in the treatment of drinking water and that BEGAC appeared to be a more advanced treatment system based on earlier operating experiences with other biological processes. The scope of this final report was thus extended to include discussion of other European biological drinking water treatment methods. But because of the complex problems of removing organic chemicals, our primary emphasis during the study phase remained on ozone and GAC systems.

Seven European drinking water treatment plants were visited that currently use ozone/GAC processing. Table 1 summarizes pertinent parameters dealing with the status of BEGAC processing at each plant. In most of these plants, criteria for reactivating the GAC have not yet been specified.

Literature Search and Review

Many papers were obtained from persons and institutions visited during the June 1978 survey. In addition, two technical conferences contributed timely, pertinent papers. One conference (Oxidation Techniques in Water Treatment) was held in Karlsruhe, Federal Republic of Germany during September 1978, and the other (Adsorption From the Aqueous Phase) was part of the 176th Annual Meeting of the American Chemical Society held in Miami Beach, Florida, also in September 1978. A search of the published literature yielded many applicable papers. Results of this literature review are interwoven throughout the report.

Results

1. The primary responsibility of a drinking water producer is to provide drinking water safe from harmful pathogenic microorganisms. To this end, water supply utilities of the United States have sought to preclude the growth of all types of microorganisms within the water treatment system. But in other countries, some water utilities intentionally incorporate biological processes into their water treatment systems to reduce the levels of dissolved organics and still maintain the microbiologically safe quality of the finished waters

2. The treatment of drinking water by the application of biological processes is not new. Biological activity is one of the processes in the slow sand filter, which was a key treatment step of early water treatment facilities, but which is generally considered obsolete in contemporary U.S. practice. But biological treatment in many forms is an important process in many European drinking water treatment systems. Examples of biological treatment of drinking water include the following.

- River sand bank filtration
- Surface storage (reservoirs)
- Gravity clarification
- Coarse media biological reactors
- Fluidized bed nitrification
- Biologically active filter media

Table 1. Water Treatment Plants Visited That Use Ozone/GAC Treatment

Plant	*Location*	*Type of Plant*	*Date Ozone Installed*	*Primary Purpose of Ozone*	*Date GAC Installed*	*Frequency of Reactivation*	*GAC Reactivation Criteria*
la Chapelle	*Rouen, France*	*Municipally owned, privately operated*	*1977*	*Preozonation for Mn + organics oxidation; post-GAC ozonation for disinfection*	*1977*	*BEGAC operating since Jan. 1977 without reactivation*	*Not yet defined*
Morsang-sur-Seine	*Villabé, France*	*Privately owned & operated*	*1970*	*Organics oxidation + disinfection*	*ca. 1975*	*BEGAC pilot unit in operation since 1977. Ran 1 yr w/o reactivation*	*Not yet defined*
Kralingen	*Rotterdam, The Netherlands*	*Municipal waterworks*	*1977*	*Disinfection + organics oxidation*	*1977*	*GAC had not operated long enough to have developed biological activity*	*Breakthrough of THM's*
Dohne	*Mülheim, FRG*	*Municipal waterworks*	*April 1977*	*Preozonation for flocculation aid; secondary ozonation for disinfection*	*Nov. 1977*	*BEGAC system operating since Nov. 1977 without reactivation*	*Not yet established. Old process (which included breakpoint chlorination): breakthrough of TOC1.*
Holthausen & Flehe	*Düsseldorf, FRG*	*Municipal waterworks*	*1954*	*Fe & Mn oxidation + organics oxidation*	*mid-1960's*	*5 to 6 months*	*When TOC1 adsorption front reaches lower GAC quadrants*
Hardhof (Lengg & Moos)	*Zürich, Switzerland*	*Municipal waterworks*	*1975*	*Disinfection/, viruses, organics oxidation*	*1975*	*every 2 to 3 years*	*When COD levels in GAC effluents increase*

- Biologically enhanced granular activated carbon (BEGAC)
- Ground passage of treated water

3. The incorporation of biological treatment steps into water treatment processes offers the following prospective benefits in water treatment:

- Reduction in the level of dissolved organic materials
- Lower oxidant (chlorine, chlorine dioxide, or ozone) demand
- Reduced operational costs
- Reduced levels of bacterial regrowths in distribution systems

4. BEGAC can be defined as the sequential unit processes (Figure 1) consisting of:

a. Oxygenation by aeration, oxygen injection, or chemical oxidation
b. Sand, anthracite, or multi-media filtration
c. Optional reoxygenation or reaeration and
d. GAC adsorption

This combination of three processes (chemical oxidation, adsorption, and biochemical oxidation) can remove ammonia and some (but not all) soluble organic substances from drinking water.

5. Dissolved organic materials in drinking water can be classified as follows:

1. Biodegradable, adsorbable by GAC
2. Biodegradable, nonadsorbable by GAC
3. Nonbiodegradable, adsorbable by GAC
4. Nonbiodegradable, nonadsorbable by GAC

Although these categories are simplified for the purpose of discussing treatment of dilute water streams, they provide a framework for postulating mechanisms by which BEGAC probably functions.

6. Both the filtration media and GAC provide supports for the biomass, which uses soluble organics and ammonia as substrates. The application of strong oxidants such as ozone to a raw water stream being treated can change the chemical nature of the dissolved organic materials. Strong oxidants can convert some (but rarely all) nonbiodegradable materials into biodegradable materials. Biochemical decomposition of organic nutrients adsorbed by the high surface area in GAC has been claimed to restore a portion of the sites to again become available for adsorption. Thus one objective of preoxidation is to couple adsorption with biological degradation.

7. The porous structure of GAC presents an ideal medium for proliferation of attached biological growth (fixed film biological growth). Both biomass and substrate are retained by the GAC—the biomass on the outer surface, and adsorbed organics in the micropores.

8. Bacteria are too large to fit into the micropores; thus they become attached to GAC media only on the outer surfaces and in the larger macropores near the outer surface that are sufficiently large to house them. As a result, only 1 to 2 percent of the total surface area available for adsorption of dissolved organics is used by the bacteria. This amount of bacterial growth is not sufficient to interfere with normal adsorption processes unless it becomes too dense and physically blocks the passages from the outer surfaces into the micropores.

9. Bacterial growths build up rapidly in GAC media. Those species that consume mainly carbonaceous organic materials appear to attain their maximum concentrations within 24 to 48 hr after virgin or reactivated GAC is placed in service. Nitrogen-converting bacteria take longer to build up to their equilibrium concentrations (30 to 90 days),

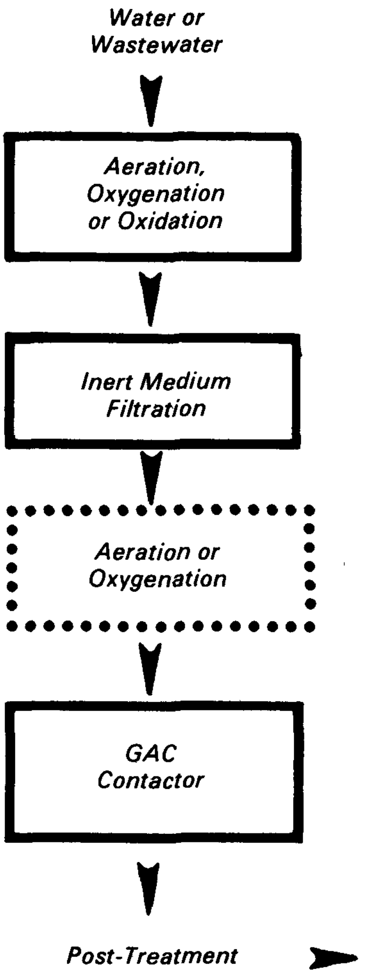

Figure 1. *Block diagram of the Biological Activated Carbon Process.*

but low levels of ammonia are converted to nitrate within a few days after fresh or reactivated GAC is placed into service.

10. Operational water treatment plants using BEGAC processes demonstrate that regeneration cycles of GAC adsorbers can be extended if a large proportion of the soluble organics entering the GAC system are biodegradable and if essential conditions (like minimum dissolved oxygen levels) are maintained. No single BEGAC system design is as yet universally accepted. So even though data exist from a number of plants, the design engineer will still have to establish design criteria for BEGAC processing in pilot plant studies. Clearly, however, designs can be developed under certain conditions to take advantage of extended GAC operational life by enhanced biological activity on both the filter media and the GAC.

11. Extension of the operational life of GAC adsorbers depends on the criteria set for reactivation. In European drinking water treatment plants using chemical preoxidation followed by GAC adsorption, these reactivation criteria have not yet been standardized from plant to plant. In those plants that have been in operation the longest using ozone followed by GAC (the three Düsseldorf plants), GAC is reactivated when the chlorinated organics adsorption front (measured as dissolved organic chlorine) reaches the lower quadrant of the GAC. Other European plants monitor levels of dissolved organic carbon, permanganate demand, UV absorption, turbidity, and taste and odor in the GAC effluents. If any of these levels increase suddenly and significantly, the GAC may be reactivated.

12. Several microbiological studies have demonstrated that the predominant microorganisms in the GAC media and in the water leaving the BEGAC system are typical soil and water bacteria. Pathogenic bacteria entering a properly designed and operated BEGAC system have been shown to be unable to compete with the predominant microorganisms present, and therefore the pathogenic species die off. Further study is required to confirm the presence or absence of harmful endotoxins. Only low dosages of post-disinfectant have been shown to be necessary to achieve the prerequisite levels of bacteriological quality of the treated water being discharged to the water distribution system.

13. Decisions to install GAC should not be based solely on the benefits to be gained from BEGAC. Rather, the decision to use GAC to remove specific organic materials should be made first. Once the decision to install GAC has been made, careful consideration should be given to extending the operational life and improving the overall organic removal process performance of the GAC by enhancing biological activity in this medium.

14. Reactivation criteria for BEGAC should be the same as those for GAC, and they should be based on the particular dissolved organic materials present in the raw water.

15. BEGAC processing will not provide any significant advantages over GAC adsorption when the dissolved organics to be removed are nonbiodegradable and cannot be made biodegradable even by chemical oxidation with ozone. Exemplary materials of this type include many of the halogenated organic compounds produced prechlorination of raw waters.

16. BEGAC systems have replaced breakpoint chlorination processes in several new and old European drinking water treatment plants. This process change has provided the advantage of avoiding production of halogenated organic materials during the early stages of the treatment process. Once halogenated organics have been synthesized, they can be removed by GAC adsorption, but only with short bed or column lifetimes. In addition, replacement of prechlorination with BEGAC systems has also produced higher quality finished water with respect to dissolved organics, ammonia, turbidity levels, and post-disinfectant (chlorine, chlorine dioxide, or ozone) demands.

17. In European water treatment plants, chemical preoxidation with ozone applied before sand, anthracite, or dual-media filtration units followed by GAC adsorption has resulted in extending the times between backwashing in each medium by a factor of about 2. Nevertheless, authorities at the Dohne plant in Mülheim have found it necessary to backwash biologically operating filters and GAC adsorbers at no greater intervals than 3 days to ensure the absence of nematodes.

18. One older European plant (the Dohne plant in Mülheim, Federal Republic of Germany) replaced breakpoint chlorination with BEGAC in 1977 at no increase in annual operating costs, including allowances for annualized capital costs.

19. When retrofitting BEGAC systems into existing drinking water treatment plants as post-adsorbers (after sand or other media filtration), provision should be made to incorporate air scouring into the backwash cycles of both the filtration and GAC media.

20. Biodegradable organic materials generally are polar and less tightly held by GAC upon adsorption. Nonbiodegradable organics tend to be nonpolar (for example, many of the halogenated organic compounds produced upon prechlorination). Some of these nonpolar, nonbiodegradable organic materials can be adsorbed to a higher degree and held more tightly by GAC. Because of these differences, some halogenated organic compounds are able to displace less strongly adsorbed polar organic materials from GAC surfaces by the process of desorption. As a result, even

though a BEGAC adsorber may be operating at biological equilibrium and may appear to be saturated with respect to adsorption of biodegradable organic materials, it may still be capable of adsorbing nonpolar, nonbiodegradable organics that are present. In such instances, reactivation could be delayed until the nonpolar, nonbiodegradable materials begin to break through the GAC medium.

21. A screening test can be conducted to determine whether BEGAC adsorption will benefit a specific raw water supply. This test shows the amount of biodegradation that can occur in the raw water supply. A sample of the raw water is ozonized with low utilized ozone dosages (1 to 10 mg/L), and the amount of biodegradable material present is compared with that of the nonozonized raw water. If oxidation of the organic materials with ozone does not increase the rate of biodegradation, then it can be concluded that BEGAC will not show any advantages over GAC for that water supply. On the other hand, if the rate of biodegradation is increased by low-level ozonation, biological enhancement of GAC should provide performance advantages. The extent of such improvements must be determined for each raw water supply to assess whether these process improvements can justify the increased costs for chemical preoxidation, preoxygenation, or preaeration.

22. Ozonation costs have been estimated for a hypothetical 50-mgd drinking water treatment plant that has installed GAC columns with empty bed contact times of 9 min and reactivation times of once every 2 months. If preoxidation with 2 mg/L of applied ozone dosage will extend the GAC reactivation time to 6 months, the costs associated with installing the required ozonation equipment are balanced by the savings resulting from GAC reactivation. Further extension of the GAC reactivation time (to 2 and 3 years, as currently occurs in some European drinking water treatment plants using BAC processes) will provide additional savings in operating costs.

Recommendations

1. Various biological water treatment processes should be investigated as to their applicability for the treatment of drinking water. Investigations should include the use of GAC as well as other adsorptive or inert media. Such studies should be conducted on systems that do not use initial breakpoint chlorination and, ideally, on systems with no prechlorination.

2. The nonpathogenic nature of bacteria should be confirmed in biologically active GAC media and in the effluents from such media.

3. The endotoxins produced by these microorganisms should be identified, and their toxicological significance should be determined.

4. Studies should be conducted to confirm the nature of the operative mechanisms occurring with BEGAC (i.e., adsorption/desorption versus apparent biological regeneration).

5. More detailed operating information should be obtained at selected European plants, including the Rouen plant in France, the Dohne, Düsseldorf, and Schierstein plants in the Federal Republic of Germany, and the Kralingen plant in Rotterdam, the Netherlands. Such information would include characteristics of influent and BEGAC media effluents about TOC, COD, DOC, UV absorption, TOC1, ammonia, etc. The specific parameters used at each operating plant should be determined to ascertain when the GAC must be reactivated.

6. Determinations should be made of the operational costs and treatment consequences of doing away with prechlorination in drinking water treatment plants (for example, modification of filter bottoms to allow for air scouring and the necessity for more frequent backwashing). Prototype U.S. plants should be operated in both modes (chlorination versus preoxidation by other means) over a 1-year cycle (minimum).

7. A variety of raw water sources should be screened to determine the applicability of biological treatment processes. Raw waters should be categorized according to the biodegradability of their organic components before and after preoxidation.

8. European water treatment operating practices should be evaluated without regard to the use of GAC with preoxidation.

9. Biological processes for nitrification of ammonia should be demonstrated as possible replacements for breakpoint chlorination.

10. The use of oxidants other than ozone should be studied for the preoxidation step. Candidate oxidants other than ozone include H_2O_2, $KMnO_4$, UV (plus air or oxygen), $C1O_2$ (free of excess chlorine), and NH_2C1 (free of excess chlorine).

11. Studies should be made of factors affecting bacterial breakthrough in BEGAC adsorbers. (Such breakthroughs have been reported in studies conducted at the Schierstein, Federal Republic of Germany drinking water treatment plant after 3 years of use.) Bacterial monitoring should possibly be considered for BEGAC systems.

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The complete report, entitled "Biological Processes in the Treatment of Municipal Water Supplies," (Order No. PB 82-199 704; Cost: $31.50, subject to change) will be available only from:

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