Comparison of advanced disinfecting methods for municipal wastewater reuse in agriculture

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Abstract The experimental investigation carried out jointly by Italian and Israeli teams within a 3-year research project funded by the European Commission on alternative disinfection methods for reusing municipal wastewater in agriculture is described. Disinfection effectiveness of UV rays, ozone (O3) and peracetic acid (PAA) at pilot scale and hydrogen peroxide (H2O2) plus silver (Ag+) or copper (Cu+2) at laboratory scale was compared for tertiary municipal effluents. Pilot plant results indicated that the well known California limit (2 CFU/100ml of Total Coliforms) was effectively met with reasonable UV doses of 100–160 mWs/cm2 or with exceedingly high PAA dosage (400 PPM); the related WHO microbial guideline (1,000 CFU/100 ml of Fecal Coliforms) was easily met with all three disinfectants (UV, PAA, O3); maximum log-inactivation values were ≥5 for UV and PAA and ≤4 for O3. Laboratory results with tertiary effluents demonstrated that Ag+ was able to inactivate target bacteria (E. coli-B, E. coli-K12), while H2O2 was more effective than Ag+ against MS-2 phages. Copper (250 µg/l) had no bactericidal effect but possessed an appreciable virucidal effect. When hydrogen peroxide and copper were combined, a pronounced increase in both bactericidal and virucidal effects was obtained.

Keywords Disinfection; copper; hydrogen peroxide; ozone; peracetic acid; silver; UV rays; wastewater reuse

Introduction Agriculture reuse of reclaimed municipal wastewater may contribute significantly to global management of water resources in semiarid regions although it requires very stringent microbial standards achievable only by advanced disinfection (Asano, 1998). Chlorination has traditionally been the most widely used disinfection method due to its ease of application, flexibility and low cost. After the discovery (Rook, 1974) that chlorination produces harmful chloro-organic disinfection by-products (DBP), however, the increased awareness to possible health and environmental impacts of chlorine has promoted a worldwide search for effective toxicologically safe alternative disinfectants, which remains a challenge for the scientific community (WEF, 1998).

To that aim, a 3-year (1995-1998) R&D project linking research teams from Israel, Malta, Morocco, Spain, UK and Italy, supported in part by the Commission of the European Community, has been carried out on technical and health care aspects of wastewater treatment for agriculture reuse. Italian and Israeli teams were jointly involved in studying the performance of advanced disinfection methods at pilot and laboratory scale, respectively. In particular, the investigation in Italy was aimed at comparing ultraviolet rays (UV), peracetic acid (PAA) and ozone (O3) disinfection in terms of pathogens (bacteria and protozoa) inactivation efficiency, DBP formation and cost evaluation using a 100 m3/h pilot plant fully equipped for treating secondary or tertiary municipal effluents by such methods (Liberti et al., 1998, 1999, in press). During laboratory scale investigations in Israel, tertiary municipal effluents were disinfected with hydrogen peroxide (H2O2)+silver (Ag+) or copper (Cu+2) ions. The mixtures were evaluated for the inactivation of target microorganisms as bacteria (E. coli-B, E. coli-K12) and bacteriophage (MS-2). Substantial efforts were
made to assess the die off kinetics of such microorganisms and to better understand possible
toxicity mechanisms of the individual agents and their combinations (Pedahzur et al., 1995,
1997).

This paper reports the results obtained with the tested disinfection methods on differently
treated municipal effluents by reference to the Italian microbial standard (2 CFU/100 ml
of Total Coliforms based on the well known Wastewater Reclamation Criteria of the State
of California, 1978) as well as to the WHO (1989) guideline (1,000 CFU/100 ml of Fecal
Coliforms) for unrestricted wastewater reuse in agriculture. The laboratory tests of the
Israeli team on inactivation of tertiary effluents are also reported.

Materials and method
Pilot investigation was carried out from July 1996 to January 1998 on a 100 m³/h demon-
stration pilot plant purposely designed, built and operated at West Bari (southern Italy)
municipal wastewater treatment plant (3,000 m³/h) to compare the performance of the three
disinfectants (UV, PAA, O₃) with three effluents of increasing quality, namely II, CL and F.
The pilot plant used during the investigation has been described in detail elsewhere (Liberti
and Notarnicola, 1999). Unchlorinated secondary (II, following secondary sedimentation)
and clarified (CL, submitted also to post-precipitation with aluminium polychloride) efflu-
ents were drawn directly from the West Bari plant. The clarified-filtered effluent (F) was
obtained by sand filtration of CL feed with a pressure filter. A 5 m³ vessel was used for batch
disinfection with the PAA mixture Oxymaster (PAA 15.5 %, H₂O₂ 22.8%, acetic acid 17%,
by Solvay Interox, Livorno, I). UV disinfection was carried out in a non-contact UV appara-
tus (mod. 600 L Super, maximum flow rate 140 m³/h, by UVT, Taranto, Italy) where the
water flow split between parallel Teflon tubes surrounded externally by low pressure Hg
vapor lamps. Disinfection with O₃ was carried out with an industrial system (mod. NFW
410, maximum production rate 445 g O₃/h, by Cillichemie, Milan, Italy) where the feed was
added with O₃ through an ejector and a hydrokinetic mixer, then entered a 5 m³ closed reac-
tion tower. O₃ was generated from air by high-tension electric discharge in a production
unit. Under the investigated experimental conditions, UV dose ranged from 30 to 430
mWs/cm², applied ozone dosage (transferred only in part to the feed) from 7 to 15 ppm with
contact time ≤15 min and PAA dosage from 1 to 500 ppm with contact time ≤60 min.
Operation procedures for each disinfectant were described in detail elsewhere (Liberti et
al., 1998, 1999, in press); analytical procedures were according to Standard Methods

Laboratory inactivation studies were conducted in Israel with tertiary effluents obtained
from the Dan region wastewater treatment plant. Typical effluent quality was: BOD
<0.5 mg/l, TOC 5.4 mg/l, COD 6.6 mg/l, pH 8.2 and turbidity 1.3 NTU. The effluents were
steam sterilized prior to use. Tertiary effluents were seeded with the target microorganisms
(\textit{E. coli B}, \textit{E. coli K12} and MS-2 phage) to achieve the desired concentration (typically
5×10⁵ CFU or PFU/ml, respectively). Hydrogen peroxide (HP), silver ions (Ag⁺), and cop-
per (Cu²⁺) were used as disinfectants. Methods for cultivation and enumeration of the
microorganisms, and the experimental inactivation tests procedures are presented else-
where (Pedhazur et al., 1995, 1997; Pedhazur, 1997; Barnea, 1998).

Results and discussion
Pilot scale investigation
During the pilot plant investigation, the three feeds (II, CL, F) were carefully monitored
with particular reference to the parameters more likely to affect disinfection effectiveness
(Table 1).
Figure 1 summarizes the most relevant results concerning the inactivation effectiveness of the given disinfectant on various feeds. These data indicate that, under the experimental conditions investigated, the Italian standard for unrestricted reuse of municipal wastewater in agriculture (2 CFU/100 ml of Total Coliforms) was effectively achieved only with UV disinfection of the best (F, clarified-filtered) or intermediate (CL, clarified) quality feed with 100 and 160 mWs/cm² UV dose, respectively, in agreement with similar investigations (Snider et al., 1991; Awad et al., 1993). Corresponding operation and maintenance (O&M) costs amounted to 17 and 35 Euro/1,000 m³, respectively. Furthermore, the limit of 2 CFU/100 ml of Total Coliforms was also achieved with PAA on F feed only with exceedingly high dosage (400 ppm) and 20 min contact time, i.e., under economically unbearable conditions (2,580 Euro/1,000 m³) according to Mandra et al. (1996).

The less stringent WHO guideline (1,000 CFU/100ml of Fecal Coliforms), indeed, was easily achieved by all three disinfectants, in particular with 30 mWs/cm² UV dose on F feed (O&M costs of 5 Euro/1,000 m³), 10 ppm of PAA for 30 min contact time on F feed (65 Euro/1,000 m³), 7 and 15 ppm of O₃ for 5 and 10 min on F and CL feeds, respectively (17 and 37 Euro/1000 m³). In addition, PAA and O₃ dosages of 50 and 15 ppm respectively for 5 min contact time permitted to achieve the interesting Total Coliforms value of 100 CFU/100 ml treating F feed. Finally, a very appealing result (≤10 CFU/100 ml of Total Coliforms) was also obtained with the poorest quality feed (II) at the maximum UV dose investigated (430 mWs/cm²).

UV physical disinfection, as expected, showed extremely fast kinetics (contact time ≤20 sec) compared with O₃ or PAA chemical disinfection. As can be seen in Figure 2, both chemical disinfectants showed a sharp initial increase of inactivation rate reaching almost completion after approximately 5 min in case of O₃ and a further increase of ≤1 log after 60 min in case of PAA. Log-inactivation values ≥5 were obtained with UV and PAA, compared with ≤4 with O₃.

Laboratory scale investigation

Silver ions and hydrogen peroxide. The activity of HP, silver and their combination against E. coli B and MS-2 phages in tertiary effluents as a function of experimental pH and temperature is presented in Figures 3a and 3b. The results clearly show that regardless of the pH or temperature, the bactericidal activity at an exposure time of 2 hours (against E. coli B) of HP at the applied concentrations was low. The virucidal activity of HP (against MS-2 phages) was more pronounced. It was not substantially effected by the temperature but decreased at pH=9.0. The decrease in activity is probably a result of the increased decomposition of HP at elevated pH. The bactericidal activity of silver ions at the concentrations tested was relatively low. Nevertheless, it increased substantially with the increase in temperature and pH. The virucidal activity of silver was low and was not effected by changes in the temperature or pH. When combined the bactericidal activity of HP and silver was

Table 1. Main characteristics of secondary (II), clarified (CL) and clarified-filtered (F) feeds

<table>
<thead>
<tr>
<th>Parameter</th>
<th>II</th>
<th>CL</th>
<th>F</th>
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<tbody>
<tr>
<td>Turbidity (NTU)</td>
<td>ave 9</td>
<td>5.1</td>
<td>29.3</td>
</tr>
<tr>
<td></td>
<td>min 5</td>
<td>11</td>
<td>6</td>
</tr>
<tr>
<td>Total suspended solids (mg/l)</td>
<td>ave 20</td>
<td>13</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>min 11</td>
<td>6</td>
<td>5</td>
</tr>
<tr>
<td>Total dissolved organic carbon (mg/l)</td>
<td>ave 13</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>min 5</td>
<td>8</td>
<td>4</td>
</tr>
<tr>
<td>254 nm Transmittance (%)</td>
<td>ave 56</td>
<td>55</td>
<td>66</td>
</tr>
<tr>
<td></td>
<td>min 44</td>
<td>55</td>
<td>66</td>
</tr>
<tr>
<td>Total Coliforms 1000 (CFU/100ml)</td>
<td>ave 1710</td>
<td>983</td>
<td>387</td>
</tr>
<tr>
<td></td>
<td>min 8</td>
<td>0.43</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td>max 630</td>
<td>4550</td>
<td>1600</td>
</tr>
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additive and increased with pH and temperature (in accordance with the increase in the activity of both agents). The virucidal activity of the combination decreased (in comparison with the activity of HP alone) and was indifferent to pH and temperature changes. The bactericidal effect of the combination of HP and silver was also conducted on wild type E. coli K-12 at 24°C and pH=7, and the results show a similar trend to those obtained with E. coli B (not presented).

Copper ions and hydrogen peroxide. In an additional set of experiments, the efficacy of HP, copper and their combinations on E. coli B and MS-2 phage was evaluated. The experiments were performed in phosphate buffer and in tertiary effluents. The results obtained in phosphate buffer displayed low bactericidal efficacy of HP (30 mg/l) and a more pronounced virucidal effect. Copper (250 µg/l) was not bactericidal but possessed an appreciable virucidal effect. When combined, HP and copper exhibited a pronounced increase in
both bactericidal and virucidal effects (not presented) (Barnea, 1998). In the phosphate buffer a reduction of 3.5 logs (against \textit{E. coli B}) and 5.2 logs (against MS-2 phage) was obtained at pH=7.0 after an exposure of 2 hours at room temperature. Applying the same HP and copper concentrations, at the same pH and temperature in tertiary effluents, a pronounced decrease in the combined efficacy (resulting in about only 1 log reduction) was achieved that was similar to the activity of HP (30 ppm) and silver (30 ppb).

Conclusions
The pilot- and lab-scale investigation carried out during a 3-year joint research project permitted the comparison of the performance of alternative disinfectants (UV rays, peracetic acid, ozone, hydrogen peroxide, silver and copper ions) to chlorination for municipal wastewater reuse in agriculture. On the basis of the experimental results obtained by treating high quality tertiary effluents, the following conclusions can be summarized.

- The stringent microbial standard for unrestricted reuse of municipal wastewater in agriculture (2 CFU/100 ml for Total Coliforms) was affordably achieved with either \textit{clarified} (CL) or \textit{clarified-filtered} (F) feeds using reasonable UV doses (160 and 100 mWs/cm$^2$, respectively). Similar results required exceedingly high doses (400 ppm and 20 min) of peracetic acid and were never achieved with ozone.
- The corresponding WHO guideline (1,000 CFU/100 ml for Fecal Coliforms) was easily achieved with all three disinfectants (UV: 30 mWs/cm$^2$, PAA: 10 ppm, 30 min, \textit{O$_3$}: 7 ppm, 5 min) on F feed.
- Total Coliforms log-inactivation values $\geq$5 for UV and PAA and $\leq$4 for \textit{O$_3$} were obtained.
- The bactericidal activity of silver, hydrogen peroxide and their combination in phosphate buffer was relatively low and possessed slow disinfection kinetics. A further decrease in activity, dominated by the diminished activity of silver, was found in tertiary effluent of high quality. This is probably due to complexation of Ag$^+$ by organic ligands present.
- The combination of hydrogen peroxide and copper (in phosphate buffer) resulted in an increased efficacy against both bacteria and viruses. Nevertheless, when applied to tertiary effluents, a pronounced decrease in activity was recorded; this is probably due to the complexation of copper by organic matter.
- Our findings suggest that the combined HP and silver disinfectant may be applied as a secondary, long acting residual disinfectant for high quality effluents.
• A 3 logs reduction using 30 ppm hydrogen peroxide together with 30 ppb silver ions required 77 minutes for *E. coli B* and 802 minutes exposure for MS-2. In comparison, it required 15 minutes using 1 ppm Cl₂ for *E. coli B* and 2 minutes for MS-2.

**Acknowledgements**

This work was partly supported by the European Commission under the Avicenne 1994 Initiative with the contract No. AVI-CT94-0010 “Advanced disinfection and health-care aspects of wastewater reclamation and reuse in agriculture in Mediterranean regions”, the US-EPA and Swissterill Water Purification Ltd., Israel. The authors wish to acknowledge the devoted work done by Y. Tchorsh, at the Division of Environmental Sciences, Hebrew University of Jerusalem, Israel.

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