Nuclear Decontamination Technology Evaluation to Address Contamination of a Municipal Water System

J. McFee, J. Langsted, M. Young
Shaw Environmental & Infrastructure
9201 East Dry Creek Rd. Centennial, CO 80112
USA

J. Porco
Michael Baker Jr., Inc
2929 N. Central Ave., Phoenix AZ 85012
USA

E. Day
Pell Resources Co.
11450 Huntsman Dr, Manassas, VA 20112
USA

ABSTRACT

The US Environmental Protection Agency (EPA) and US Department of Homeland Security (DHS) are considering the impact and recovery from contamination of municipal water systems, including intentional contamination of those systems. Industrial chemicals, biological agents, drugs, pesticides, chemical warfare agents, and radionuclides all could be introduced into a municipal water system to create detrimental health effects and disrupt a community. Although unintentional, the 1993 cryptosporidium contamination of the Milwaukee WS water system resulted in 100 fatalities and disrupted the city for weeks.

Shaw Environmental and Infrastructure Inc, (Shaw), as a subcontractor on a DHS contract with Michael Baker Jr., Inc., was responsible for evaluation of the impact and recovery from radionuclide contamination in a municipal water system distribution system. Shaw was tasked to develop a matrix of nuclear industry decontamination technologies and evaluate applicability to municipal water systems. Shaw expanded the evaluation to include decontamination methods commonly used in the drinking water supply. The matrix compared all technologies for implementability, effectiveness, and cost. To address the very broad range of contaminants and contamination scenarios, Shaw bounded the problem by identification of specific contaminant release scenario(s) for specific water system architecture(s).

A decontamination technology matrix was developed containing fifty-nine decontamination technologies potentially applicable to the water distribution system piping, pumps, tanks, associated equipment, and/or contaminated water. Qualitatively, the majority of the nuclear industry decontamination technologies were eliminated from consideration due to implementability concerns. However, inclusion of the municipal water system technologies supported recommendations that combined the most effective approaches in both industries.

INVESTIGATION

The contractual objective of this project was to prepare a matrix of processes that could be used to decontaminate municipal water systems (MWS), materials, and components from a radionuclide contamination event. The overall approach involved:

- Identify and evaluate unique contamination/decontamination issues for municipal water systems when considering radionuclide contamination;
- Develop performance/selection criteria to first screen and then rate the technologies;
- Develop a list of decontamination technologies;
- Perform a preliminary screening of the technologies to remove obvious non-candidate technologies from further consideration; and
- Apply the selection criteria to narrow the list to the most applicable options.

Figure 1 graphically depicts the selection process.

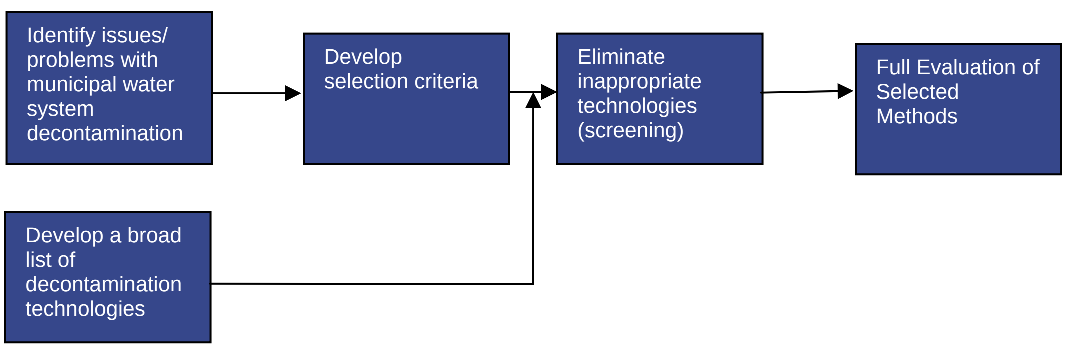

Figure 1; Graphical decontamination technology selection process

Identify Issues/Problems with Municipal Water System Decontamination

In the nuclear industry decontamination technology selection/application has matured to nearly an intuitive selection of applicable options and a cost evaluation of the appropriate technologies. However, there are application issues in municipal water systems that are not incurred in typical nuclear decontamination problems.

Municipal water systems have approaches and techniques to deal with organic contamination, usually by adding agents such as chloramines or hypochlorite to or kill organisms. Since radioactive contamination is inorganic in nature, it is largely unaffected by these typical MWS organic oxidation/disinfection technologies.

A second issue is that radionuclide contamination can be dangerous at very low mass concentrations. Table 1 lists drinking water concentration limits in activity units and mass units for four selected radionuclides under two possible cleanup standards; the EPA drinking water standards and the DHS Proposed Protective Action Guides (1).

Table 1, Drinking Water Concentration Limits for Four Radionuclides and Two Cleanup Standards

Contaminant	EPA Drinking Water Standard		DHS Proposed Protective Action Guide	
	Activity Concentration	Mass Concentration	Activity Concentration	Mass Concentration
Cs-137	7.40 Bq/l (200 pCi/l)	2.3E-06 µg/l	510 Bq/l (1.38E+04 pCi/l)	1.59E-04 µg/l
Co-60	3.70 Bq/l (100 pCi/l)	9.1E-08 µg/l	965 Bq/l (2.61E+04 pCi/l)	2.37E-05 µg/l
Sr-90	0.3 Bq/l (8 pCi/l)	6.0E-08 µg/l	194 Bq/l (5.25E+03 pCi/l)	3.75E-05 µg/l
Am-241	0.55 Bq/l 15 pCi/l	4.4E-06 µg/l	11.8 Bq/l/ (3.19E+02 pCi/l)	9.38E-05 µg/l

One significant feature of Table 1 is the extremely low mass levels required for compliance with either EPA's drinking water standards or the DHS Proposed Protective Action Guides. Removal of inorganic contaminants from water down to these very low levels is an engineering challenge. Decontaminating a complete water system to these levels requires extensive testing and engineering. This challenge is complicated by the fact that the contaminants are contained in a water "matrix" containing chemically similar non-radioactive elements or minerals.

Other problems, issues, and deployment challenges that must be considered include:

- Highly toxic or carcinogenic decontamination agents are unacceptable in drinking water systems.
- Some nuclear industry methods require application conditions that cannot be achieved in drinking water systems, such as elevated high temperatures.
- Leakage in large complex municipal water systems as high as 10% is common. Groundwater contamination is a potential secondary issue.
- The highly uncertain nature of the radionuclide contamination event complicates pre-identification of the decontamination approach. The variability includes the specific radionuclide, the total activity introduced, the size of the distribution system, the mode of introduction, the time and method of detection, and the chemical form of the radionuclide.
- The need to ensure the integrity of drinking water system components and infrastructure both during and after decontamination. During decontamination, maintenance of water pressure for fire protection must be considered. Secondly, depressurization of a water system for any reason can result in inadvertent contamination for other sources.
- Complexities in MWS system configurations present unique decontamination problems. The configurations included pumping stations, supply tanks, water mains, and miles of distribution lines. Distribution system piping buried under roads and buildings with numerous crud traps that are difficult to locate. Complicating factors/variables of the distribution system include pipe type; size; initial water quality; connection type; and the degree of sediment, scale, and biofilm in the system (2).

- Multiple materials of construction need to be taken into consideration. Cast iron and steel pipe tend to have greater problems with biofilm, scale, and tuberculation (mounds of corrosion products). Concrete pipes are usually coated. Newer plastic piping systems are less likely to scale.
- Public reaction, cooperation, compliance, and community acceptance of the proposed decontamination approach adds complexity to an already difficult and challenging task.
- Non-zero contamination of the water with very low (legal) levels of naturally occurring radionuclides may not be understood by the public when debating cleanup standards.
- Detection of the contamination in the inaccessible piping distribution systems is problematic
- The potential for adsorption/absorption or physical incorporation of these contaminants in the biofilm, pipe walls, sediment, and pipe scale has substantial impact on decontamination requirements (2). Chemical/physical interaction issues include:
    1. The radionuclide may react or adsorb on the scale, pipe material, biofilm, and sediments within a system. It is conceivable that the radionuclides bind to these surfaces within a short distance of introduction, or progress miles downstream.
    2. Alternatively, precipitated radionuclide compounds may settle out and slowly dissolve back into the system.
    3. The radiation may kill the absorbing biofilm and be released back into the system as radioactive organic material.

Develop Selection Criteria

These issues, along with more typical technology evaluations, were used to develop technology selection criteria. For simplicity, the criteria were grouped into the categories of effectiveness, implementability and cost. The criteria were subsequently reviewed with municipal water system representatives to confirm viability.

Effectiveness Criterion**:**

- The decontamination method must be capable of providing water quality meeting either EPA drinking water standards or DHS protective action guides, as applicable to the situation. This is a difficult criterion recognizing the very low mass concentrations in the standards shown in Table 1 and the variety of materials to be decontaminated. This criterion should be considered a go/no-go criterion. However, due to the lack of implementation data, it remained a qualitative evaluation.
- Long-term effectiveness
    1. No long-term or delayed health effects are predicted due to desorption or leaching back into the drinking water at an unacceptable rate or concentrations after decontamination; and
    2. The potential for groundwater contamination resulting from mobilized contaminants leaking from the system is minimized.

Implementability Criterion:

- Decontamination chemicals are not toxic in applied quantities (2). This is a go/no go criterion;

- Risk to workers implementing the decontamination is minimized, including the safe handling of decontamination materials;
- Decontamination agents (chemicals) are readily available or stockpiled in sufficient quantities;
- Method uses readily available equipment and can be applied across the broad system or applied by multiple crews in targeted areas;
- Method enables a rapid return to a minimal but acceptable level of service in accordance with the Safe Drinking Water Act limits;
- Method does not require elevated temperatures to be effective (for piping only);
- Method does not require unreasonable contact (soaking) time to be effective;
- Method does not degrade or erode distribution system integrity;
- Safety impact due to depressurized system is minimized or avoided; and
- Techniques for the management of decontamination flushings and residuals are acceptable, and the method does not produce an excessive amount of contaminated secondary waste.

Cost Criteron:

- Cost of materials (decontamination agents) is manageable;
- Cost of implementation (labor and equipment) is manageable; and
- Cost of waste management (principally flushings and associated debris, but also personnel protective equipment and other contaminated materials) and disposal is manageable

Develop a broad list of decontamination technologies

A list of known decontamination technologies was developed from Shaw corporate knowledge and several literature references (3, 2). The complete list includes chemical and mechanical decontamination techniques known from Shaw decontamination projects as well as those identified in nuclear journals and references. In addition, typical municipal water system technologies were added, since they are known to be implementable and cost effective.

The following are decontamination methods commonly applied to municipal water systems.

- Conventional flushing is widely used in the MWS to respond to water quality issues. It is the practice of simply opening one or more fire hydrants and allowing water to flow until sediment or other contaminants are removed (2).
- Unidirectional flushing is an improvement on conventional flushing in that valves are closed and hydrants opened in a systematic fashion that maximizes water velocities above those incurred in normal operation (2).
- Air scouring uses pulses of water and air to clean pipelines. This technique requires a variety of special equipment not normally in the MWS inventory, including a compressor, air filters, air cooler, baffle box, filter socks, and tanks (2).
- Pigging (Polyurethane and Swab)uses water pressure to push a swab or polyurethane cylinder, bullet-shaped, or dumb bell-shaped pig through the line. Pigging removes layers of sediments, biofilm, and soft scale and hard scale. Pigging of four- to six-inch lines can be

launched and recovered through most existing fire hydrants without significant additional equipment. Larger pipes and pigs require launch and receiver components. These may be present in large transmission lines, but for most pipelines it would require excavating down to the pipe, cutting into the pipe and installing the launcher, and then excavating at the end of the pipe to cut into the pipe and install a pig receiver. Pigging becomes more effective as pipe size increases (2).

- Abrasive pigging is the same as pigging except that it uses a pig with abrasive materials such as Velcro, carbide straps, plastic brushes, or wire brushes embedded in the shell of the pig to more aggressively remove harder scales. (2)
- Chemical decontamination is effective on removal of contaminants from the pipe wall, sediment, biofilm, friable scale, and hard scale. Unlike the mechanical methods which remove sediment, biofilm, friable scale, and hard scale, chemical decontamination attacks each matrix and dissolves the radionuclides. The effectiveness of this method and the problems associated with its use depend on the chemical selected. Ellison indicates that chemical decontamination is considered very effective against biofilm, friable scale, and hard scale, but does not recommend chemical decontamination for sediment, since it does not actually remove the sediment (2).
- Pipeline relining options include spray-on concrete linings as well as insertion of linings of a smaller size. Although this is not a "decontamination" process per se, it is listed here as an expeditious option for consideration by a utility. This option may be particularly interesting in the case of low levels of contamination or as a final assurance that radionuclide desorption is precluded in the system.

Initial Screening

During this step in the process, ineffective or inappropriate decontamination methods for a radionuclide contamination event were qualitatively eliminated from further consideration. In this screening stage, the general characteristics of effectiveness and implementability were considered, as opposed to the specific quantifiable evaluation. Chemicals used in nuclear decontamination that were classified as toxic or poisonous, based on MSDS information, were excluded as unimplementable.

A subset of toxicity includes chemicals that have been shown to contaminate groundwater. Since municipal water systems are known to incur significant leakage, groundwater protection becomes a secondary, but real issue (4). Complexing agents have been shown to keep contaminants mobilized through soil conditions without breaking down, and ultimately impacting groundwater (5, 6). They form strong bonds with some radionuclides and have been shown to survive both aerobic and anaerobic digestion at wastewater treatment plants. This poses a waste disposal problem as well as a groundwater hazard. Therefore technologies using the complexants such as ethylenediaminetetraacetic acid (EDTA), diethylentriaminopentacetic acid (DTPA), or oxyethylidenediphosphonic acid (OEDPA), were excluded as toxic and therefore unimplementable.

The required temperature of application screened out some nuclear decontamination methods (7). Some nuclear decontamination methods require application at temperatures up to 100 degrees Centigrade (ºC) in order that rate kinetics are practicable. Significant temperature elevation is not implementable in drinking water distribution system pipes, valves and pumps. Elevated temperature is possible in tanks and this criterion was not applied to those applications.

The last screening category was demonstrated effectiveness for decontamination of systems in terms of size and characteristics. Some nuclear decontamination methods are intended for application within gloveboxes or hot cells, or are techniques that could not be applied to large systems (3, 7). Several nuclear technologies were removed from consideration based on lack of demonstrated effectiveness applicable to large systems (3).

Proposed Decontamination System for Treatment of Water.

Treatment of contaminated water was also considered in this DHS project. The magnitude of the water treatment requirements and the extent of the treatment requirements led to the conclusion that general recommendations would be difficult. Therefore, the treatment technology evaluation was based on an assumed contamination scenario. This approach differs somewhat from the decontamination of equipment, but represents evolution of the project and recognition of the uniqueness of radionuclide contamination.

The selected scenario resulted in 605 million liters to be treated at an average radionuclide concentration of 2.1E+06 Bq/l (5.7E+7 pCi/l). This concentration is 10,000 times the EPA release limits to a publically owned treatment works. No single water treatment process is likely to economically achieve this cleanup. Therefore, a multi-process treatment train was developed based on Shaw experience for the most cost effective use of individual treatment processes. That process is shown in Figure 2, and includes;

1. The first element of the process is a multi-step filtration process.
2. A reverse osmosis system;
3. Ion exchange media for a final polishing step;
4. An evaporator is included to reduce the volume of the reverse osmosis rejects stream; and
5. The evaporator bottoms contain all the non-volatile water minerals and are therefore cemented for disposal as radioactive waste.

Based on the assumed scenario and the resulting engineering requirements, evaluation of water treatment technologies against each other was not attempted. A complete "system" is likely to be required to adequately address a radionuclide attack. Clearly, if the attack targeted a smaller facility, or resulted in lower concentration decontamination solutions, reverse osmosis alone may be adequate. Similarly, if pigging only were used on a small target, an evaporator could be the technology of choice.

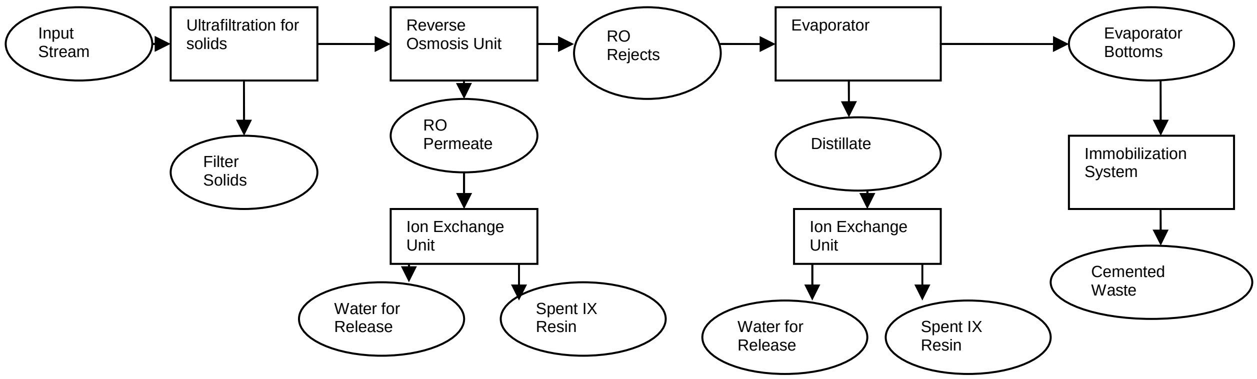

Figure 2, Proposed treatment train for decontamination water treatment

Full Evaluation of Surviving Technologies;

Decontamination methods that passed the initial screening were evaluated to determine their relative effectiveness on the MWS using each radionuclide of concern. The methods commonly used by MWS to decontaminate pipe may be quite effective at responding to a radionuclide contamination event on a MWS and by their common usage are readily implementable with relatively low cost and complexity. In order to use decontamination effectiveness studies already conducted by the American Water Works Association Research Foundation on MWS pipelines, the decontamination methods were regrouped from simplest to most complex/expensive, using the method names and types familiar to the MWS operators. (2)

Most of the nuclear decontamination methods applicable to the piping, valves, and pumps of a distribution system can be considered a single subset of MWS methods (chemical decontamination). The evaluation of chemical decontamination includes all the chemical decontamination methods listed in Table 2 and compares effectiveness based on radionuclide chemical interactions. Each method was assessed for its effectiveness against radionuclides in sediment, biofilm, and soft and hard scale. While it is possible that bare pipe materials are present, it is very likely it will be coated with some level of biofilm and scale; therefore, decontamination effectiveness on bare pipe was not considered a prime screening parameter (2). The cost of each method versus the other methods was also evaluated based on the AWWARF reference. Finally, the cost of each method versus its effectiveness was evaluated.

Table 2 shows the complete technology list indicating the effectiveness and implementability assessment for each technology according to application to the piping system, tanks or for contaminated water cleanup. The recommended technologies are identified.

Decontamination Recommendations

This search for effective and implementable decontamination technologies has provided an opportunity to evaluate available technologies in the current environment. The uncertainties in the nature of any contamination and the unknowns of chemical effects in the distribution system make definitive technology selections tenuous. However, this evaluation supports the preliminary recommendations that the chemical decontamination agent, citric acid, used in the nuclear industry and commonly used MWS mechanical technologies (swabbing or abrasive pigging) would appear to be appropriate methods for piping. Citric acid, a very common decontamination chemical used in the nuclear industry, is at the top of the list of decontamination chemicals for the following reasons:

- It forms strong aqueous complexes with many expected radionuclide contaminants;
- It has a very good track record for decontamination at nuclear sites (3,8);
- It is biodegradable and it is recognized as safe for use in food, so getting approval for use in a drinking water system should be easier (9);
- Public acceptance should be much easier than with the other chemicals; and
- It has very good handling properties.

Acetic acid is the next best choice. It has properties that are similar to citric acid in terms of effectiveness except that it is does not form complexes with americium. Another drawback is that acetic acid has a strong unpleasant odor. However, an advantage with acetic acid is that americium is more soluble in concentrations of acetic acid that would be used for decontamination.

Phosphoric acid is third on the list. It forms aqueous complexes with americium and strontium.

A wider range of methods appears effective for decontamination of tanks/equipment. The many mechanical and chemical methods used by the nuclear decontamination industry have a proven track record at nuclide decontamination.

Table 2 Final Technology Applicability and Ranking

Decontamination Method	Nuclear Decon Method	Piping Implementable	Piping Technically Effective	Piping Cost Effective	Tanks Implementable	Tanks Technically Effective	Tanks Cost Effective	Water Implementable	Water Technically Effective	Water Cost Effective	Recommended
Chemical Decontamination Options											
CAN-DECON	●										
CAN-DEREM	●										
VNIPIET (Russian) electrochemical	●										
Complexing agents	●										
Ethylenediaminetetraacetic acid (EDTA), complexing agent	●										
LANL electrochemical	●										
CORPEX	●										
Laser ablation	●										
Anaerobic bio-decon	●				●						
Strippable coatings; as low as reasonably achievable (ALARA) 1146, GP-RDM, Tech Sol, Stripcoat TLC, Pentek 603/604 (and others)	●				●						
Acid salts	●	●			●						
ADA electrochemical	●	●			●						
Bases/alkaline salts	●	●			●						
Sulfuric acid	●	●			●						

Decontamination Method	Nuclear Decon Method	Piping Implementable	Piping Technically Effective	Piping Cost Effective	Tanks Implementable	Tanks Technically Effective	Tanks Cost Effective	Water Implementable	Water Technically Effective	Water Cost Effective	Recommended
Decontamination for Decommissioning (DfD)	●				●	●					
Fantastic/Spic & Span/Alconox/ commercial detergents	●	●			●						
Hydrochloric acid	●	●			●						
Hypochlorus acid		●			●			●			
Isosaccharinate (actinides) on concrete	●	●			●						
Low oxidation metal ion (LOMI)	●	●			●						
Nitric acid	●	●			●						
CORD	●				●	●	●				
CITROX	●				●	●	●				
Fluoroboric acid	●				●	●	●				
Cerric nitrate	●				●	●	●				
Nitric acid permanganate (NP)	●				●	●	●				
NITROX	●				●	●	●				
Oxalate-peronic-gluconic acid (OPG)	●				●	●	●				
Textract/RadPro	●				●	●	●				
Organic acids	●	●	●	●	●	●	●				
Alkaline permanganate	●	●	●	●	●	●	●				
Phosphoric acid	▲	▲	▲	▲	▲	▲	▲				YES
Acetic acid		▲	▲	▲	▲	▲	▲				YES
Citric acid	▲	▲	▲	▲	▲	▲	▲				YES

CONCLUSION

Decontamination of a municipal water system from a radionuclide contamination event will require a rapid, but in-depth, review of the decontamination options available and the constraints imposed by the particular water system age, materials, client base, and contamination extent. General recommendations on criteria for decontamination technology selection as well as likely decontamination agents are made. However, these general recommendations must be evaluated carefully for applicability in a real contamination event.

If the contamination event is identified quickly the issue of treatment of the contaminated water must be addressed. In either case, large volumes of contaminated water are likely from the decontamination and flushing processes. Those volumes will require complex and expensive treatment for release.

REFERENCES

1. Federal Register (FR), January 3, 2006 "Protective Action Guides for Radiological Dispersal Device (RDD) and Improvised Nuclear Device (IND) Incidents" US Department of Homeland Security.
2. Ellison, D. *et al.* 2003. *Investigation of Piping Cleaning Methods,* AWWA Research Foundation. 2003.
3. International Atomic Energy Agency (IAEA), 1999. "State of the Art Technology for Decontamination and Dismantling of Nuclear Facilities," IAEA Technical Reports Series No. 395.
4. Los Alamos National Laboratory (LANL), 2002. *Analysis of Municipal Water Systems for Assessment of Radiological Dispersal Threats or Terrorism,* LA-UR-02-2121. Rees, B. et al. April.
5. Cokesa, E., H. J. Knackmuss, and P.G. Rieger. 2004. "Biodegradation of All Stereoisomers of the EDTA Substitute Iminodisuccinate by Agrobacterium tumefaciens BY6 Requires an Epimerase and a Stereoselective C-N Lyase." Applied Environmental Microbiology: 70(7): July 2004.
6. Nowack, B., F. G., Kari, and H. G. Krüger. 2001. "The Remobilization of Metals From Iron Oxides and Sediments By Metal-EDTA Complexes." Water, Air, and Soil Pollution, 2001.
7. American Society of Mechanical Engineers (ASME), Draft Nuclear Facility Decommissioning and Decontamination Handbook, Chapter 17 Decontamination,
8. International Atomic Energy Agency (IAEA), 2002. "Application of Ion Exchange Processes for the Treatment of Radioactive Waste and Management of Spent Ion Exchangers," IAEA Technical Reports Series No. 408.
9. United Nations Environment Program (UNEP), 2001. "Citric Acid," CAS No: 77-92-9, SIDS Initial Assessment Report, January 2001