Environmental Science & Technology

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Comparative Assessment of the Environmental Sustainability of Existing and Emerging Perchlorate Treatment Technologies for Drinking Water

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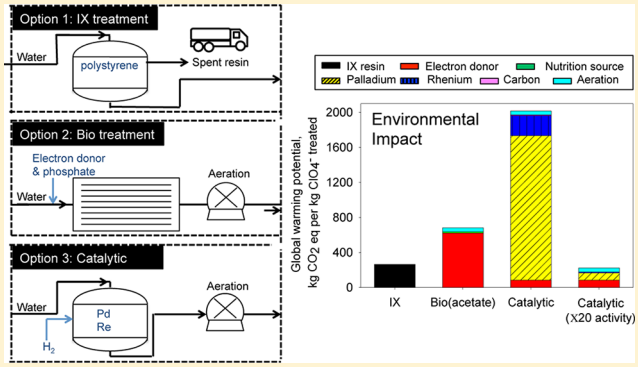
Supporting Information

ABSTRACT: Environmental impacts of conventional and emerging perchlorate drinking water treatment technologies were assessed using life cycle assessment (LCA). Comparison of two ion exchange (IX) technologies (i.e., nonselective IX with periodic regeneration using brines and perchlorate-selective IX without regeneration) at an existing plant shows that brine is the dominant contributor for nonselective IX, which shows higher impact than perchlorate-selective IX. Resource consumption during the operational phase comprises >80% of the total impacts. Having identified consumables as the driving force behind environmental impacts, the relative environmental sustainability of IX, biological treatment, and catalytic reduction technologies are compared more generally using consumable inputs. The analysis indicates that the environmental impacts of heterotrophic biological treatment are 2−5 times more sensitive to influent conditions (i.e., nitrate/oxygen concentration) and are 3−14 times higher compared to IX. However, autotrophic biological treatment is most environmentally beneficial among all. Catalytic treatment using carbon-supported Re−Pd has a higher (ca. 4600 times) impact than others, but is within 0.9−30 times the impact of IX with a newly developed ligand-complexed Re−Pd catalyst formulation. This suggests catalytic reduction can be competitive with increased activity. Our assessment shows that while IX is an environmentally competitive, emerging technologies also show great promise from an environmental sustainability perspective.



INTRODUCTION

Perchlorate (ClO_4^-) is used in the manufacture of rocket propellant, explosives, and fireworks, and the contamination of water resources results from improper use and disposal of these materials.[1,2] It also occurs naturally in arid regions and potash ore within the United States.[3] Its properties (anionic, water-soluble, chemically inert) make perchlorate both highly mobile and persistent in groundwater and surface water.[4] A recent U.S. EPA survey reported that perchlorate has been detected in more than 50 surface water sources and more than 60 groundwater sources used by drinking water utilities in the United States.[5] Drinking water contamination by perchlorate raises human health concerns because perchlorate interferes with iodine uptake by the thyroid, preventing normal endocrine system function.[6] Because of potential risks associated with perchlorate consumption, state regulatory standards have been implemented in California (6 μg/L) and Massachusetts (2 μg/L), and the U.S. EPA recently announced plans to set a federal regulatory standard.[7−10] Most of the utilities with perchlorate-contaminated source water serve from 10 000 to 100 000 people, and 35 of the surveyed utilities provide water to >100 000 people. As the drinking water industry invests in technology to overcome this challenge, there is a need to understand the relative sustainability of and trade-offs among perchlorate treatment technologies in terms of treatment efficacy, cost, and life cycle environmental impacts.

Among available technologies, ion exchange (IX) is most commonly implemented in the United States.[11] There are over two dozen plants that use IX systems to treat perchlorate in drinking water in California and Massachusetts, where perchlorate is regulated. Commercial IX processes are capable of meeting the regulatory levels with short reactor residence times.[12] Previously, IX resins used for perchlorate treatment were not highly selective for perchlorate (hereafter referred to as nonselective IX), and require frequent resin regeneration because of uptake of nontarget ions that are typically much more abundant in source waters (e.g., NO_3^-, SO_4^{2-}, HCO_3^-). Resins are regenerated by flushing resin beds with concentrated salt (NaCl) brines. The spent brines contain high concentrations of salt, perchlorate, and other nontarget anions, and may require further treatment before discharging.[12] Although

Received: October 20, 2012
Revised: February 26, 2013
Accepted: March 13, 2013
Published: March 13, 2013

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regeneration prolongs the resin lifetime, concern has been raised regarding negative environmental impacts associated with brine generation and its disposal. More recently, IX resins that are highly selective for perchlorate (hereafter referred to as perchlorate-selective IX) have been developed, resulting in much longer contaminant breakthrough times from resin beds.[13] However, the high selectivity for perchlorate also makes resin regeneration with brines ineffective; thus, spent resins are typically disposed after a single use, often by off-site incineration. Although nonselective IX is still common—particularly where treatment of nitrate is also required—many utilities treating perchlorate-contaminated source waters have adopted perchlorate-selective IX primarily due to its lower cost and ease of operation and maintenance.[14] However, the environmental implications of this transition have never been assessed, and it is unknown whether this reduction in cost comes at a price for the global environment.

Biological treatment is another option for perchlorate removal, where anaerobic microorganisms reduce perchlorate to chloride using organic carbon (e.g., acetate, ethanol) or hydrogen as the electron donor.[15] Several pilot-scale studies have shown the effectiveness of biological treatment under typical groundwater conditions and have demonstrated cost-competitiveness of the processes.[16−18] However, implementation of biological treatment still faces challenges in overcoming public opposition to the use of biological processes for drinking water treatment and concerns about pathogen introduction. Hence, biological treatment is still considered an emerging technology for drinking water. Although biological treatment processes are generally perceived to be "greener" and more sustainable than physical/chemical technologies,[19] a formal comparative assessment of life cycle environmental impacts of biological versus IX treatment technologies for perchlorate have not been reported.

Recently, chemical reduction of perchlorate has emerged as a third treatment option. In the presence of carbon-supported Re-Pd bimetallic catalysts, reduction of perchlorate to chloride was observed when hydrogen was supplied as an electron donor.[20] Its activity was found to be several orders of magnitude faster than other chemical reduction technologies for perchlorate (e.g., zerovalent iron[21,22]), and recent work demonstrates effectiveness for treating perchlorate present in waste IX regenerant brine.[22] Although it is still in the early stage of development, catalytic reduction shows promise as an alternative technology, but the environmental implications of the technology are yet to be quantified.

Given that federal regulation of perchlorate in drinking water is imminent,[10] it is important to elucidate the trade-offs among treatment technologies. Although economic costs are often a basis for comparison, no studies to date have objectively compared the relative environmental sustainability of existing and emerging perchlorate treatment technologies. The goal of this study is to evaluate the life cycle environmental impacts of conventional and emerging perchlorate treatment technologies using life cycle assessment (LCA).[23−25] LCA has been applied to a wide range of processes in the water industry, from specific treatment technologies to entire wastewater treatment plants.[26−28] Technologies evaluated here include nonselective and perchlorate-selective IX, heterotrophic and autotrophic biological treatment, and catalytic treatment. The IX treatment processes are first evaluated for a specific water treatment plant where these technologies have been in use, and results are then generalized to compare all perchlorate treatment technologies. Specific objectives are to (i) quantify the relative contribution of each material and process to each environmental impact category for each treatment technology, (ii) elucidate the trade-offs among treatment technologies from an environmental perspective, and (iii) identify key opportunities for each technology to advance environmental sustainability and reduce treatment costs through future research.

■ MATERIALS AND METHODS

System Description. This study is presented in two parts. First, the environmental implications of choosing a perchlorate-selective IX system over a nonselective IX system with brine regeneration for perchlorate treatment are evaluated (herein referred to as **IX Selectivity Impact Assessment**). The two conventional systems are compared based on information obtained from an existing drinking water treatment plant in southern California. The facility treats approximately 3.6 million gallons-per-day (MGD) of groundwater that is distributed to approximately 2500 residents for domestic purposes, as well as for commercial, industrial, and agricultural purposes. The influent ClO_4^- concentration ranges from 20 to 60 μg/L. The facility recently switched from a nonselective IX system to a perchlorate-selective IX system. Figure 1 shows a schematic of the treatment plant with system components of nonselective IX and perchlorate-selective IX. The nonselective IX system includes brine regeneration, resulting in a more complex configuration with additional components compared to perchlorate-selective IX. Detailed descriptions of the two IX systems and their subsystems are provided in the Supporting Information (SI).

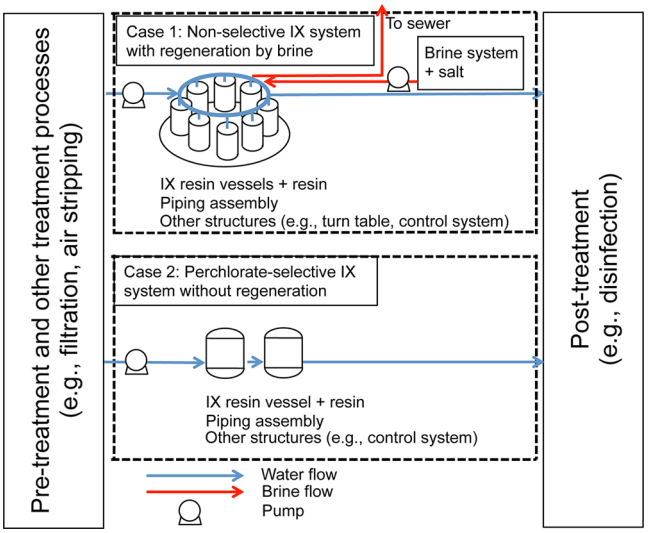

Figure 1. Schematic of alternative IX treatment systems used at the California utility; box with dashed line indicates system boundary. Detailed description for each component can be found in the SI.

Using the perchlorate-selective IX as a new benchmark, the second part of this study (herein referred to as **Emerging Technology Assessment**) evaluates emerging treatment technologies: biological reduction and catalytic treatment. To generalize results, perchlorate-selective IX is compared to these technologies based on the environmental impacts of consumable inputs normalized to the mass of perchlorate treated. The presumption—supported by LCA results from the first part of the study—is that consumables are the dominant

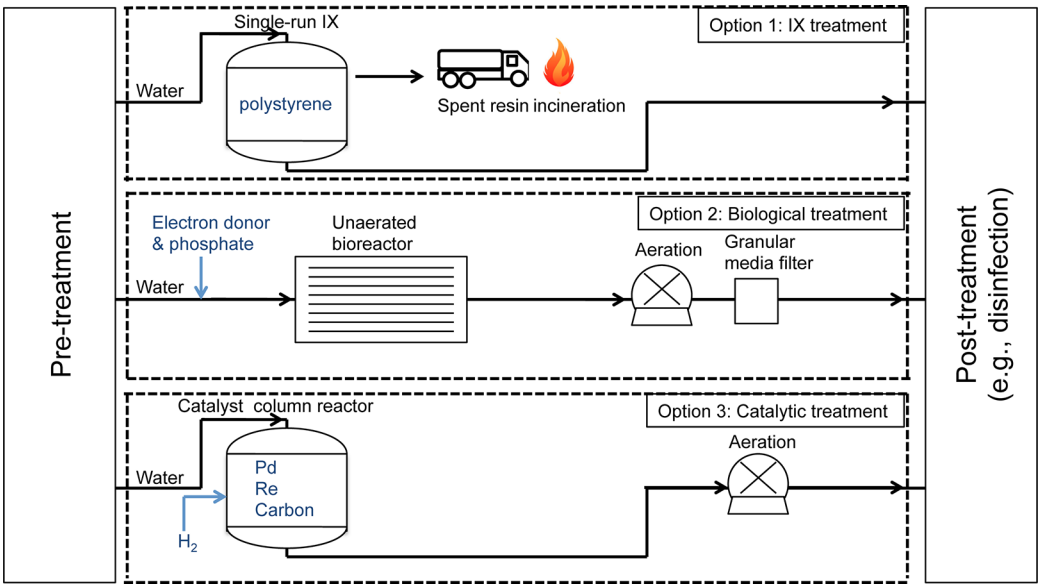

Figure 2. Schematic of (1) perchlorate-selective IX, (2) biological reduction, and (3) catalytic reduction for perchlorate treatment technologies; box with dashed line indicates system boundary.

contributors to each process' life cycle environmental impacts, and therefore, assessment of their environmental impacts can typify that of the entire system. This assumption is also supported by results reported previously by others studying environmental impacts of water and wastewater treatment technologies.[26,28−30] While this study focused on comparison of the aforementioned perchlorate-targeted treatment technologies, other treatment technologies such as reverse osmosis may be alternative options, particularly if a new plant is being constructed to treat multiple classes of emerging contaminants.

The process flow diagrams for perchlorate-selective IX, biological reduction, and catalytic reduction are shown in Figure 2. For perchlorate-selective IX, pretreated water passes through IX resin packed in a cylindrical vessel, and resin is replaced when its adsorption capacity is reached. Spent resins are transported off-site and incinerated.

For biological reduction, electron donor (e.g., acetate, ethanol, hydrogen) and nutrients such as phosphate are added to support growth. The water then passes through an unaerated bioreactor where microorganisms reduce dissolved oxygen, nitrate, and perchlorate. The treated water is filtered via biologically active carbon and a sand filter to remove biomass and biodegradable organic materials. Subsequently, water is aerated to reintroduce dissolved oxygen. Kinetic parameters for biological reduction were obtained from the literature.[31,32]

For catalytic reduction, water passes through a column packed with carbon-supported Re-Pd catalysts, where hydrogen is added to promote the same reactions discussed for biological reduction processes. Like biological reduction, the treated water is then aerated. Kinetic parameters for catalytic reduction were obtained from our previous study.[33]

System Boundary. The goal of this LCA was to provide a comparative assessment of the life cycle environmental impacts of alternative perchlorate treatment technologies. As such, the system boundary for this study primarily considers the construction and operation of system components that are directly related to perchlorate treatment and does not include other on- and off-site drinking water treatment processes that would be common to all alternatives. For instance, raw water is pretreated and passed through a booster pump to increase head pressure prior to perchlorate treatment, and disinfected after perchlorate treatment. These processes are the same for all perchlorate treatment technologies, and are thus outside of the system boundary. Incineration of the perchlorate-selective resin, although occurring off site, was included within the system boundary because it is unique to one technology option and is considered integral to the overall life cycle impacts of the consumable resin that is assumed to be replaced annually.

For the IX Selectivity Impact Assessment (part 1), the functional unit is removal of perchlorate from 3.6 MGD of influent water for 10 years, where influent perchlorate concentration is 50 μg/L and effluent perchlorate concentration is below 6 μg/L. The functional unit was chosen based on the duration that nonselective IX was used in this treatment plant before switching to the new perchlorate-selective IX. For the Emerging Technology Assessment (part 2), the functional unit was the removal of one kilogram of perchlorate with an influent containing 50 μg/L perchlorate and an effluent below 6 μg/L. The system boundary of the Emerging Technology Assessment was also modified to focus on consumable inputs as the driver of each technology's life cycle environmental impacts.

Life Cycle Inventory. For the IX Selectivity Impact Assessment (Part 1), the inventory lists of materials and energy for two IX systems were generated from information obtained at the California treatment plant. These include all stationary units such as vessels, pipelines, and mechanical and electrical components, in addition to consumable inputs such as resins, chemicals, and energy use involved for the perchlorate treatment system. While construction of the IX system is not shown as a separate process, the study included the construction phase of the IX system by incorporating all raw materials and processes required for the material manufacturing into inventory compilation. For nonselective IX, detailed system schematics from the manufacturer were obtained and used. For perchlorate-selective IX, materials and their quantities were determined and scaled from those of nonselective IX systems based on their relative IX tank sizes. A description of components included in each subsystem of nonselective and

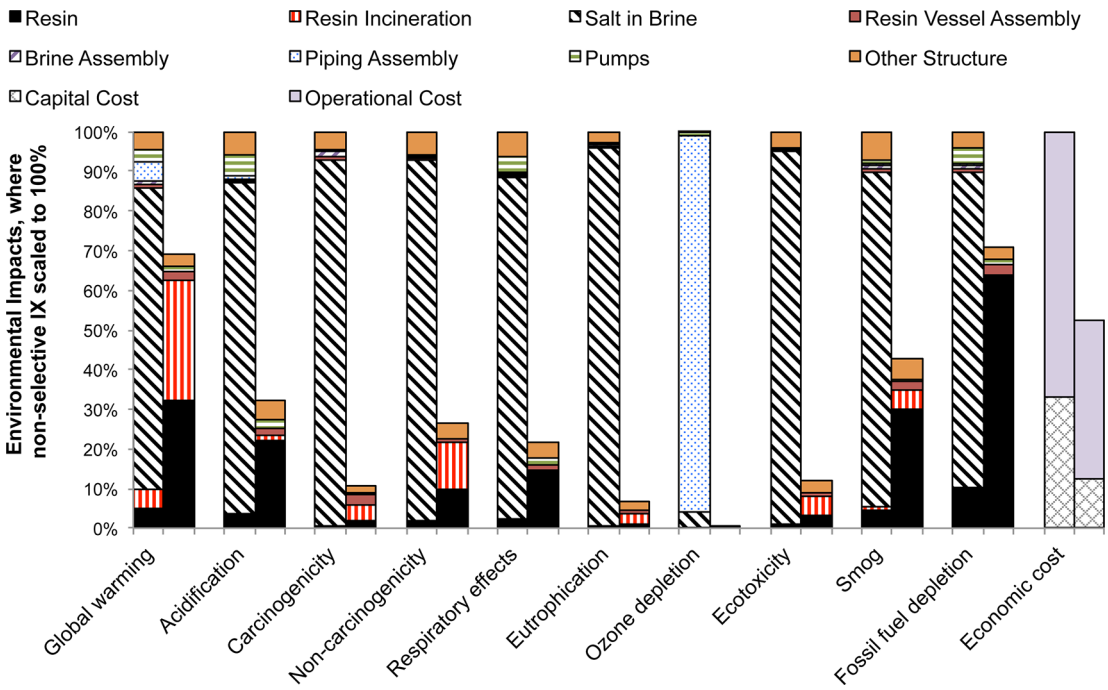

Figure 3. Comparison of environmental impacts and cost of nonselective IX with brine regeneration (left bar in each category) to perchlorate-selective IX without regeneration (right bar in each category). Note that the impacts from incineration of the spent perchlorate-selective IX resin are shown separate from other impacts associated with resin production and use.

perchlorate-selective IX and the breakdown lists of materials and their masses for two systems are provided in the SI.

For the Emerging Technology Assessment (Part 2), inventory lists were generated from consumables used during the operational phase of each treatment system. The quantities of consumables were calculated based on the information obtained from the literature and are summarized in the SI. For both the first and second parts of this study, the life cycle inventory data for each material (i.e., resources and emissions associated with extraction of raw materials, manufacturing, and transportation processes) were obtained from EcoInvent database (v2.2) implemented in SimaPro (v7.3; PRé Consultants; The Netherlands).

Impact Assessment. The environmental sustainability of alternative perchlorate treatment technologies was evaluated by comparing environmental impacts of these technologies. The study was performed following the general framework for LCA established by the International Organization of Standardization (ISO).[24,25] The environmental impacts of each treatment technology were assessed using the Tool for the Reduction and Assessment of Chemical and other environmental Impacts (TRACI) v2.0,[34] which uses the following midpoint environmental impact categories: global warming (kg CO_2-equivalent), acidification (H^+ moles-eq.), human health (kg benzene-eq., kg toluene-eq., and kg $PM_{2.5}$-eq. for carcinogenicity, noncarcinogenicity, and respiratory effects, respectively), eutrophication (kg N-eq.), ozone depletion (kg CFC-11-eq.), ecotoxicity (kg 2,4-dioxane-eq.), smog formation (g NO_x-eq.), and fossil fuel depletion (MJ-surplus).

RESULTS AND DISCUSSION

Part I: IX Selectivity Impact Assessment. LCA results for the comparison of nonselective and perchlorate-selective IX treatment systems are summarized in Figure 3. Results for each impact category are normalized to absolute values of the nonselective IX system because each category uses different units and absolute scales are not comparable across categories. This is not meant to imply that the impacts in different categories are of equal magnitude or importance. Corresponding absolute values with units are provided for each category in the SI. Also shown are relative economic costs for the two systems [based on personal communications with the Calgon Carbon Corporation], separated into capital and operating costs. Actual capital and operating costs are provided in the SI.

For the nonselective IX system, salt (NaCl) used for the brine contributed over 80% of impacts in all categories, except ozone depletion. Although a large contribution from salt consumption was expected, such dominance across 9 of 10 categories was not anticipated by the authors. While the environmental impacts of salt are smaller than those of other materials (e.g., polymers used for resins) when they are compared on a mass basis, the large salt usage (9 tons/day) by the treatment plant was shown to overwhelm the impacts from all other materials combined. The major environmental impacts from salt use results principally from toxic chemicals released during the solution mining process where water is injected into a salt deposit.[35] Alternative salt sources ($CaCl_2$ and LiCl) were also evaluated, but their impacts in all categories were significantly higher than for NaCl (1- to 2-orders of magnitude higher). The large impacts from salt usage suggests that efforts to treat and recycle waste brines significantly reduce overall environmental impacts of nonselective IX systems and should be a key direction for future IX research.

For perchlorate-selective IX, production and incineration of IX resins are major contributors to environmental impacts ranging from 52% to 90% in all categories, except ozone depletion. The IX resin is mostly made of a polystyrene polymer, which contributes to the above environmental categories. The environmental impacts from the IX resin are principally attributed to compounds emitted to air during the

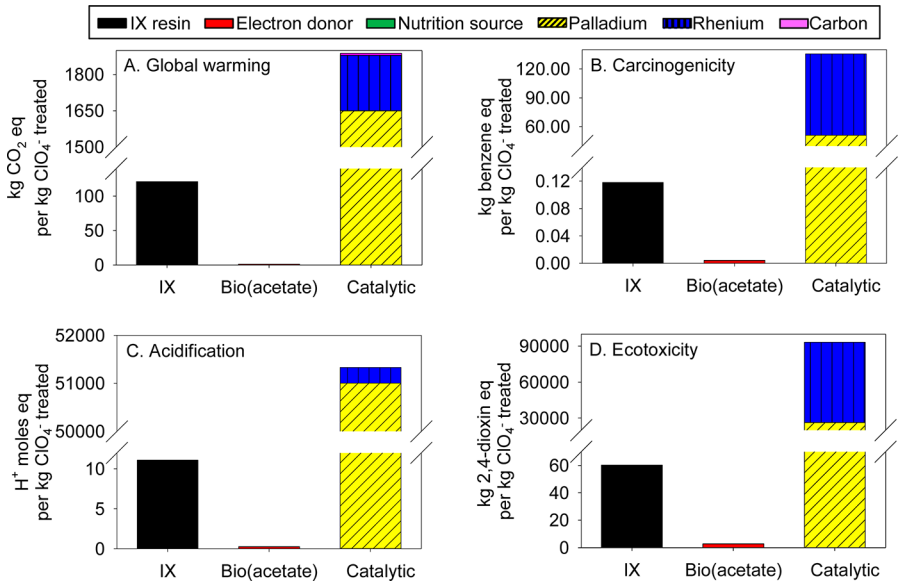

Figure 4. Environmental impacts of perchlorate-selective IX, biological reduction treatment and catalytic treatment for idealized perchlorate treatment (no other water constituents such as dissolved oxygen or oxyanions are considered).

combustion process and to water during the distillation process of polystyrene production.[36] The relatively high contribution of IX resins to the category of fossil fuel depletion (about 91%) is due to energy intensive processes in the production.

For all categories, the environmental impact of the perchlorate-selective IX system is less than that of the nonselective IX system. The IX resin is the dominant contributor in most categories for the perchlorate-selective IX, as a larger amount of IX resin (approximately 7 times higher) is required since spent resins need to be disposed of and replaced annually; however, this increase in the total amount of IX resin resulted in smaller impacts than the salt (in brine solution) used for regeneration of the nonselective IX resin. In all categories except global warming and fossil fuel depletion, the impacts from the perchlorate-selective IX were less than 40% of those determined for the nonselective IX. For global warming and fossil fuel depletion categories, impacts from the perchlorate-selective IX system were approximately 70% of the nonselective IX system. In the former, production and incineration of the polymer-based resin was the dominant contributor to fossil fuel consumption. When evaluation time is extended to 20 years, the changes in relative contribution of salt use and resin were less than 5%. The dominance of impacts from salt use and resin did not change since dominant contributors for all components are consumable inputs (e.g., salt, resin) from operational phase and their magnitude increases proportionally to the evaluation time. Comparison of capital and operating costs (in Figure 3) also shows the advantage of the perchlorate-selective IX system over the nonselective IX system, which is not surprising since this has been a primary driver for the recent industry trend toward perchlorate-selective IX treatment systems. Uncertainty values of environmental impacts of two IX systems were estimated using the Pedigree matrix[37] and Monte Carlo simulation; calculation details and results are shown in SI. Uncertainty values are small relative to differences between environmental impacts of nonselective and perchlorate selective IX.

Comparison of the two IX systems at the California treatment plant showed that perchlorate-selective IX is more environmentally sustainable than nonselective IX with brine regeneration for perchlorate removal from contaminated water. This work also identified that the dominant contributions to environmental impacts were from consumables (e.g., resins, salt) used during the operational phase (mostly above 80%). This latter conclusion is consistent with results reported by others studying environmental impacts of water and wastewater treatment technologies.[26,28−30] This shared observation has a significant implication for comparative assessments of alternative perchlorate treatment technologies: the relative environmental sustainability can be evaluated based on consumables used during the operational phase, as these consumables will likely drive life cycle environmental performance. Such approximation is especially useful for the evaluation of emerging treatment technologies for which limited (or no) full-scale data is available, but where quantities of consumable inputs can be estimated based on a fundamental understanding of the technology.

Part II: Emerging Technology Assessment. The environmental sustainability of perchlorate-selective IX and emerging perchlorate treatment technologies (i.e., heterotrophic and autotrophic biological reduction, chemical reduction using Re-Pd/C catalyst) are assessed and compared to explore the current technology landscape, as well as to identify fundamental limitations of and future research directions for each technology. Based on the result that consumables used during operational phase of IX treatment systems represent the major portion of environmental impacts (and supported by similar observations in water and wastewater studies), the relative environmental sustainability of these treatment technologies was evaluated using only consumable inputs. Both ideal (where only perchlorate was present in source water) and nonideal (where nontarget anions and alternative electron acceptors were present in source water) conditions are considered.

Idealized Perchlorate Removal. Resin or electron donor requirements were calculated for removal of only the target perchlorate under the assumption that there is no inhibition from or competition with other constituents in water (e.g.,

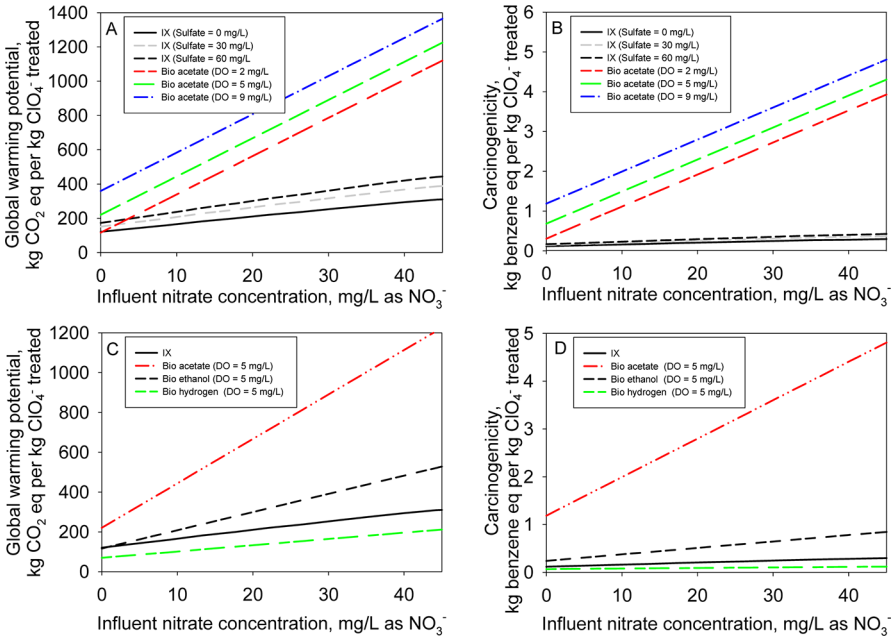

Figure 5. Sensitivity of environmental impacts of selective IX and biological reduction perchlorate treatment processes to influent nitrate concentrations and other water conditions (A, top left, and B, top right) and different electron donors (C, bottom left, and D, bottom right); environmental impact for aeration process is included for all biological reduction.

dissolved oxygen, nitrate). Figure 4 shows direct comparison of environmental impacts (global warming, human health carcinogenicity, acidification, and ecotoxicity) of perchlorate-selective IX, biological reduction using acetate as the electron donor, and catalytic reduction using hydrogen in the presence of carbon-supported Re-Pd catalysts. The quantity of consumable inputs for each technology is normalized per kg of ClO_4^- treated. Assumptions and summary of calculations are shown in SI. The results show that the environmental impacts of biological reduction are the lowest by a wide margin, followed by IX, then catalytic reduction for all impact categories. For biological reduction, the contribution from electron donor is 87−98% in these categories. Catalytic reduction results in environmental impacts 2−3 orders of magnitude greater than the other two technologies. The dominant contributions stem from catalyst metals (i.e., palladium, rhenium). For these metals, sulfidic tailings contribute to human health and ecotoxicity, and natural gas burned during the refining of palladium contributes to global warming and acidification.

Impact of Source Water Quality. Resin or electron donor requirements were calculated for removal of perchlorate, and other constituents in water. For IX, other anions (e.g., nitrate, sulfate, bicarbonate, chloride) compete with perchlorate for uptake on resin sites. For biological reduction, other electron acceptors (e.g., nitrate, sulfate, oxygen) can inhibit perchlorate reduction by microorganisms, as they are energetically more favorable. Similarly, other electron acceptors react with hydrogen and compete for reaction sites during catalytic reduction. Among these constituents, nitrate is a key constituent because it has the second highest affinity to IX resins, and it is a favorable electron acceptor because it is often present at concentrations 3−4 orders of magnitude greater than perchlorate.

Focusing on perchlorate-selective IX and biological reduction as leading alternatives (from an environmental perspective), Figure 5 shows the sensitivity of environmental impacts of these technologies to influent water constituents, especially to nitrate concentration. For biological reduction, the environmental impacts of global warming and carcinogenicity increase linearly with nitrate concentration, because electron donor requirements increase linearly with alternative electron acceptor concentrations (e.g., nitrate). The sensitivity of environmental performance to dissolved oxygen in the influent is also evaluated across the range of 2−9 mg/L, where the higher end is the saturation level. Since biological reduction removes dissolved oxygen from water, biological treatment systems must also reaerate treated waters, which result in additional environmental impacts (which were included in this analysis). For IX, the influence of nitrate and sulfate on the performance of IX was calculated based on results reported in literature.[38] Equilibrium multicomponent chromatography was used to predict the effect of nitrate and sulfate concentrations on run length to perchlorate breakthrough for perchlorate-selective resins. The change in environmental impacts due to the presence of nitrate (up to 44 mg/L, MCL level) and sulfate (up to 60 mg/L) are smaller than those of biological reduction. The results indicate that environmental impacts of treatment technologies are sensitive to common water constituents other than perchlorate, but IX is more environmentally sustainable than heterotrophic biological reduction using acetate as electron donor under typical water conditions.

For biological reduction, the effects of different electron donors (i.e., acetate, ethanol, hydrogen) on environmental performance of the system have also been evaluated. Among three common electron donors, acetate results in the highest environmental impacts followed by ethanol, then hydrogen. Acetic acid is produced by the Monsanto process,[36] in which carbon monoxide reacts with methanol at high temperature and pressure and produce carbon dioxide as a side reaction. For human health carcinogenicity, the emission of heavy metals from the disposal of sulfidic tailings is the major contributor. Ethanol for industrial use is produced from direct hydration of ethylene, where carbon dioxide is emitted, and hard coal

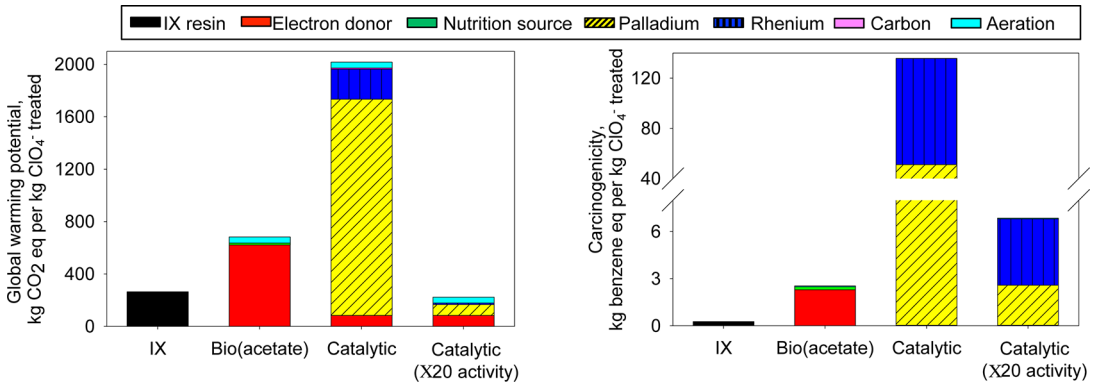

Figure 6. Environmental impacts of selective IX, biological reduction with acetate, and catalytic reduction processes treating influent water with 50 μg/L perchlorate, 20 mg/L nitrate, 30 mg/L sulfate, and 5 mg/L dissolved oxygen.

Table 1. Comparison of Environmental Impacts of Perchlorate Treatment Processes to Conventional Drinking Water Treatment Operations

					conventional treatment		% relative to conventional treatments		
impact category	unit	IX[a]	bio (acetate)[a]	catalytic[a]	ref 40[b]	ref 41[c]	IX[d]	bio (acetate)[d]	catalytic[d]
global warming	kg CO_2 eq	260	680	2000	8400	3100	3.1, 8.4	8.1, 22	23, 64
acidification	H^+ moles eq	24	170	51 000	2900	1500	0.83, 1.6	5.8, 11	1700, 3300
carcinogenics	kg benzen eq	0.26	2.5	140	33	13	0.77, 2.0	7.4, 19	410, 1000
non carcinogenics	kg toluen eq	4200	17 000	190 000	220 000	88 000	1.9, 4.8	7.5, 19	84, 214
respiratory effects	kg $PM_{2.5}$ eq	0.097	0.90	250	15	8.4	0.64, 1.1	6.0, 11	1600, 2900
eutrophication	kg N eq	0.089	1.6	87.3	31	16	0.28, 0.5	5.0, 10	280, 540
ozone depletion	kg CFC-11 eq	1.4×10^{-7}	0.00012	0.00017	0.0025	0.00017	0.005, 0.1	4.6, 69	6.9, 100
ecotoxicity	kg 2,4-D eq	130	1600	93 200	19 000	17 000	0.68, 0.8	8.3, 9.3	480, 540
smog	g NO_x eq	0.24	1.7	8.7	11	5.4	2.2, 4.4	15, 31	82, 160
fossil fuel resources	MJ surplus	440	2300	2800	14 000	4200	3.1, 10.4	17, 56	20, 66

[a]Functional unit is per kg ClO_4^- treated from influent water condition is assumed to be 50 μg/L perchlorate, 20 mg/L nitrate, 30 mg/L sulfate, and 5 mg/L dissolved oxygen. [b]Environmental impact is assessed based on operational phase inventory list of a drinking water treatment plant in Waternet, Netherland.[40] The treatment processes include raw water intake, coagulation, lake water reservoir, pumping from the reservoir, rapid sand filtration, ozonation, softening, biological activated carbon filtration, and slow sand filtration. [c]Environmental impact is assessed based on operational phase inventory list of a drinking water treatment plant in Quebec, Canada.[41] The treatment processes include coagulation-flocculation-sedimentation, granular filtration, granular activated carbon adsorption, chlorination, and corrosion control. [d]First number is relative to the environmental impact of drinking water treatment ref 40 and second number is relative to that of the treatment ref 41.

burned in industrial furnaces for ethanol production contributes to the release of carcinogenic heavy metals. Hydrogen is produced from steam reforming of fossil fuels (i.e., natural gas). The reformation process is the major source of carbon dioxide emission, which is produced as a side reaction. The mass of electron donor required to reduce 1 kg of perchlorate is 0.6 kg acetate, 0.3 kg ethanol and 0.08 kg hydrogen based on the stoichiometric ratio (stoichiometric reactions and biomass yield assumptions are shown in SI); therefore, differing consumption levels (on a mass basis) of alternative electron donors greatly contribute to the differences in their total environmental impacts to treat perchlorate. Additionally, the impacts of acetate on human health categories are approximately 1 order of magnitude higher than those of hydrogen on an equivalent mass basis. While environmental contributions of heterotrophic biological (i.e., acetate, ethanol as electron donor) reduction are larger than those of IX over the range of nitrate concentrations shown in Figure 5, autotrophic biological reduction (i.e., hydrogen as electron donor) results in a smaller environmental impact than IX, demonstrating its promise as a more environmentally sustainable technology. It is also worth noting that the environmental advantage of using hydrogen as electron donor over others can also be applied when evaluating alternative electron donors for in situ bioremediation strategies (e.g., for reductive dehalogenation of chlorinated solvents).

Figure 6 shows environmental impacts of catalytic perchlorate treatment compared to IX and biological reduction in the presence of sulfate, nitrate, and oxygen under typical concentrations found in perchlorate-contaminated drinking water sources. Catalytic reduction also requires additional electron donor for reduction of nitrate and oxygen, and this increases environmental impacts of the technology in addition to the impact from the aeration process. However, the changes in environmental impacts are relatively small (less than 7%), since metal catalysts are the dominant contributors. The environmental sustainability of catalytic perchlorate reduction can be significantly improved if catalytic activity can be enhanced. Recent research focuses on improving catalytic activity by modifying the coordination environment of rhenium metal (i.e., Re-ligand complexes).[39] These modifications result in ca. 20 times higher activity than catalysts considered in calculations presented above. The environmental impacts of catalytic reduction when activity is enhanced by 20 times (thus reducing required amounts of metals by a similar factor) are also shown in Figure 6. For global warming, the contribution from metal catalysts is reduced to 42% of the total impact; this makes the total impact of the catalyst system comparable to IX

and biological reduction. For human health, catalytic reduction still shows higher impacts than IX and biological reduction, as 98% of the impact comes from palladium and rhenium metals. Uncertainty values of environmental impacts for the conditions evaluated in Figure 6 are in the SI; they are relatively small compared to differences between environmental impacts of these treatment options. It is important to recognize that the environmental impacts of catalytic reduction for other pollutants cannot be inferred from those for perchlorate. For example, Pd-based catalytic reduction rate constants for two common pollutants, trichloroethylene and nitrate, are ca. 3800 times and ca. 40 times greater (normalized by Pd mass) than that for perchlorate. Hence, the environmental sustainability of alternative technologies for each pollutant must be analyzed individually.

Impact of New Perchlorate Regulation on Drinking Water Treatment. To study the impact of a new perchlorate regulation on existing drinking water treatment plants, the environmental impacts of perchlorate treatment technologies are compared relative to those of conventional drinking water treatment technologies in Table 1. These conventional treatments include processes such as coagulation-flocculation-sedimentation, granular media filtration, and chemical disinfection. The inventory lists of consumable inputs for these processes were obtained from literature,[40,41] and the treatment processes included in these lists are described in Table 1. Their environmental impacts were characterized in the same way as those of perchlorate treatment technologies. For perchlorate-selective IX, environmental impacts from consumable inputs are estimated to be <10% of those estimated for conventional treatment technologies. Impacts from heterotrophic biological reduction are in the range of 5−30% of conventional treatment technologies for most impact categories. Catalytic reduction shows environmental impacts that are comparable to those of conventional treatment processes, and even show higher impacts in categories of acidification, human health, eutrophication, and ecotoxicity. This suggests that for catalytic reduction to be a viable treatment option for perchlorate, its environmental impacts need to be significantly reduced. This can be achieved by enhancing catalyst activity (i.e., 50−500 $Lh^{1-}g_{Pd}{}^{-1}$ with equivalent wt% Re) as previously discussed. These results also indicate that consumables associated with new unit processes for perchlorate treatment will increase the overall environmental impacts of a treatment facility. Thus, it follows that improvement in these technologies that reduce consumable inputs (e.g., technologies enabling recycling of waste IX brines, perchlorate-selective IX resins that are regenerable, more active catalysts, use of lower-impact electron donors) can contribute measurable improvements to the sustainability of drinking water treatment operations as a whole. This finding is useful for researchers setting priorities for future technology development goals (e.g., setting benchmarks for catalyst activity). The marked effect of nontarget water constituents on the relative environmental impacts of competing perchlorate treatment processes also highlights the importance of considering site-specific source water characteristics in selecting the most sustainable treatment options to use at individual utilities.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

A table listing the summary of assumptions made in this study, detailed listing of inputs, and a table listing cost analysis of two IX systems are provided. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by NSF Chemical, Bioengineering, Environmental, and Transport Systems (CBET No. 07-30050) and U.S. EPA (RD-83517401). Its contents are solely the responsibility of the grantee and do not necessarily represent the official views of the EPA. Further, the EPA does not endorse the purchase of any commercial products or services mentioned in the publication. Charles Drewry (Calgon Carbon Corporation) is acknowledged for providing information on capital and operating expenses for IX systems. We also thank Youneng Tang (UIUC), John Shapley (UIUC), and Thomas Theis (UIC) for insightful discussion and technical feedback.

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