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Life Cycle Comparison of Environmental Emissions from Three Disposal Options for Unused Pharmaceuticals

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Supporting Information

ABSTRACT: We use life cycle assessment methodology to compare three disposal options for unused pharmaceuticals: (i) incineration after take-back to a pharmacy, (ii) wastewater treatment after toilet disposal, and (iii) landfilling or incineration after trash disposal. For each option, emissions of active pharmaceutical ingredients to the environment (API emissions) are estimated along with nine other types of emissions to air and water (non-API emissions). Under a scenario with 50% take-back to a pharmacy and 50% trash disposal, current API emissions are expected to be reduced by 93%. This is within 6% of a 100% trash disposal scenario, which achieves an 88% reduction. The 50% take-back scenario achieves a modest reduction in API emissions over a 100% trash scenario while increasing most non-API emissions by over 300%. If the 50% of unused pharmaceuticals not taken-back are toileted instead of trashed, all emissions increase relative to 100% trash disposal. Evidence suggests that 50% participation in take-back programs could be an upper bound. As a result, we recommend trash disposal for unused pharmaceuticals. A 100% trash disposal program would have similar API emissions to a take-back program with 50% participation, while also having significantly lower non-API emissions, lower financial costs, higher convenience, and higher compliance rates.

INTRODUCTION

The disposal of unused, unwanted, or expired pharmaceuticals contributes to the occurrence of active pharmaceutical ingredients (APIs) in aquatic environments,[1,2] wastewater biosolids,[3] and treated drinking water.[4] Since these compounds can have negative impacts on both human and environmental health,[2,5−8] incineration is being utilized as a way to eliminate unused pharmaceuticals as a source of APIs in the environment. "Take-back" disposal, which entails consumers transporting unused pharmaceuticals to a collection site for incineration, is increasing in popularity as an environmentally conscious means to dispose of unused pharmaceuticals. Several take-back disposal programs have been recently piloted in the United States (U.S.).[9−12]

Previous studies have investigated current practices[2] and risk management goals[13] for the disposal of unused pharmaceuticals. Other studies have evaluated the cost[14] and convenience[10] of take-back programs, as well as the public's willingness to pay[15] for and participate[16] in them. Amidst this research, the number of voluntary take-back disposal programs is increasing, and new U.S. legislation is making the national implementation of take-back programs a possibility.[17] On the other hand, the majority of environmental API emissions may arise from human and animal excretion; further, the impact of the disposal practices on non-API emissions (e.g., greenhouse gas emissions or smog forming potential) is not yet established.[18]

This study quantifies the environmental emissions of APIs as well as the emissions of other non-API substances that result from the disposal of unused pharmaceuticals. Using life cycle assessment methodology, the study compares the three disposal options illustrated in Figure 1: (i) take-back disposal where pharmaceuticals are driven to a pharmacy to be incinerated as hazardous waste; (ii) toilet disposal where pharmaceuticals are flushed down a toilet to be treated as domestic wastewater; and (iii) trash disposal where pharmaceuticals are mixed with an unpalatable item and put in the household trash to be managed as municipal solid waste (MSW), as recommended by the White House Office of National Drug Control Policy (ONDCP).[2]

METHODS

The three disposal options for unused pharmaceuticals are evaluated using a comparative life cycle assessment following the ISO 14040 framework.[19] Figure 1 lists the major steps for the three disposal options considered in this study. The functional unit is the disposal of an annually accrued mass of unused pharmaceuticals and associated packaging from U.S. households, where it is assumed that the unused pharmaceutical mass is evenly distributed across U.S. households.

The annual mass of unused pharmaceuticals is estimated as 90 million kilograms (200 million pounds).[20] This mass is represented by the 10 most commonly returned pharmaceuticals during a take-back pilot program.[12] Each of these

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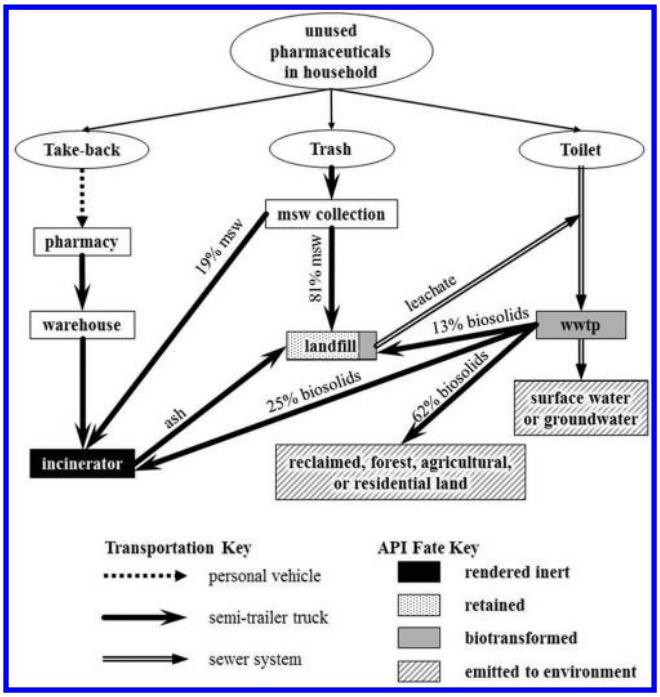

Figure 1. Waste management systems used by each disposal option, including the transportation and fate of APIs. Abbreviations: msw = municipal solid waste; wwtp = domestic wastewater treatment plant.

pharmaceuticals is assumed to have the same disposal mass. Since 2 of the 10 most commonly returned pharmaceuticals contained acetaminophen as their API, the mass of acetaminophen is estimated at 18 million kilograms while the other 8 representative APIs are estimated at 9 million kilograms each. Associated packaging waste is assumed to be 14 million kilograms, based on masses collected during a take-back pilot program.[10] Composition of the packaging material is based on general U.S. packaging waste[21] and pharmaceutical specific packaging.[22] Brief descriptions of data sources, calculations, and assumptions are provided below. Complete information is available in the Supporting Information and at http://deepblue.lib.umich.edu/ ("Unused Pharmaceutical Disposal").

Emissions Categories. In this analysis, 10 environmental emissions categories are considered, with 9 categories for non-API emissions and 1 category for emissions of APIs. Non-API emissions arise from the production, use, and/or disposal of materials, fuels, electricity, and infrastructure used in each disposal option. The production of pharmaceuticals is common across all disposal routes and is not included within the LCA system boundary. Non-API emissions are estimated using the Ecoinvent,[23] Franklin USA 1998,[24] and U.S. Life Cycle Inventory (LCI)[25] databases, with adjustments made to reflect typical U.S. transportation and energy system characteristics by substituting U.S. data for European data. The Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI)[26] is used to aggregate the hundreds of non-API emissions into the nine non-API emissions categories, based on emission equivalence calculations (e.g., global warming potential in equivalent CO_2 emissions).

The fate of APIs in this study includes the following: incineration, retention in a landfill by sorption, biotransformation by aerobic or anaerobic processes within a wastewater treatment plant (WWTP) or landfill, or emission to the environment with WWTP effluent or land-applied biosolids. Other possible API fates are assumed to be negligible. For example, the direct emission of APIs from a landfill due to leakage of leachate is assumed to be insignificant based on observed leachate collection efficiencies.[27,28] The incineration process is also assumed to be effective, so the resulting ash does not contain a significant amount of APIs. Therefore, the total API mass emitted to the environment results from the wastewater treatment of flushed pharmaceuticals (toilet disposal) and the wastewater treatment of landfill leachate containing APIs (trash disposal).

Due to uncertainty regarding biotransformation and sorption, an uncertainty analysis is performed with wide parameter ranges, shown in Table 1, to encompass the lack of precise data regarding biotransformation, desorption, sorption equilibrium, and heterogeneity of biomass and MSW. Since knowledge and data about transformation products is not currently available,[8] all biotransformed APIs are assumed to be either (i) oxidized or assimilated into biomass under aerobic conditions or (ii) fully converted into landfill gas (e.g., methane, carbon dioxide) in anaerobic environments. Other than these, if the products of biotransformed APIs are shown to be a significant health or environmental hazard, future research should be incorporated within the framework of this study.

Trash. In the trash disposal option, participants mix their pharmaceuticals with a waste item (e.g., coffee grounds) inside a low-density polyethylene (LDPE) bag and remove the

Table 1. Fates of Each Representative API in a WWTP and a Landfill with the Maximum (Max) and Minimum (Min) Values Used to Estimate Mass Percent Biotransformation and Sorption[a]

compound	WWTP					landfill			
	overall % removal	% sorption		% biotransformation		% biotransformation		% sorption	
	(ref)	min	max	min	max	min	max	min	max
acetaminophen	95%[39]	0%	82%	0%	100%	0%	55%	20%	100%
aspirin	86%[38,39]	61%	100%	0%	39%	0%	0%	75%	100%
vitamin E	100%[40]	75%	100%	0%	25%	0%	0%	75%	100%
prednisone	96%[39,42]	51%	100%	0%	49%	0%	9%	66%	100%
ibuprofen	90%[39,43]	65%	100%	0%	35%	0%	0%	75%	100%
warfarin	80%[41]	55%	100%	0%	45%	0%	0%	75%	100%
topiramate	15%[44]	0%	40%	0%	40%	0%	0%	75%	100%
etodolac	45%[44]	20%	70%	0%	50%	0%	0%	75%	100%
gabapentin	99%[39]	0%	25%	49%	100%	0%	99%	0%	100%

[a]Min and max value calculations are in the Supporting Information (section 3, page S9).

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labeling from the associated packaging as suggested in the 2007 ONDCP statement.[2] Since our model assumes all trash participants follow the ONDCP recommendation, the LDPE bag and packaging are placed in household trash and no pharmaceuticals are placed in a recycling or composting bin. The production of the LDPE bag (but not the waste item, such as coffee grounds) is included in the LCI. Garbage trucks collect this trash with MSW and, according to current U.S. MSW management, haul 19% to an incinerator and 81% to a landfill.[21]

Incineration of the plastic bag and contents produces air emissions and ash. Air emissions and the composition of ash resulting from the plastic bag and packaging are derived from LCI data. Incineration is assumed to completely oxidize APIs, and the resulting air emissions are based on each representative API's chemical formula as well as treatment of the resulting flue gas with currently available technologies.[23,29,30] Energy recovery from incineration is included in the model by offsetting electricity production. The amount of electricity offset is based on an assumed energy density of 0.535 MWh/ton combusted.[31]

The API fates in a landfill include biotransformation, sorption, or removal with the leachate. All bags are expected to lose their structure due to landfill activities (e.g., mechanical compaction) and release the enclosed APIs. Emissions arising from landfilling the plastic bag and pharmaceutical packaging are estimated with LCI data. Emissions from APIs are calculated using API properties to determine leachate and landfill gas (LFG) compositions and production quantities. The anaerobic biotransformation potential in a landfill for APIs is estimated as a fraction of the aerobic biotransformation potential (in the WWTP). This ratio is defined with an uncertainty parameter ($N_{LF/WWTP}$). The resulting LFG is assumed to be 50% methane, consistent with typical compositions.[32] According to the U.S. average, LFG is either flared to reduce harmful gaseous emissions (28%), combusted to generate electricity (31%), or directly emitted to the atmosphere (41%).[33] The model system boundary includes displaced electricity production due to LFG-based energy recovery.

Within the landfill, the masses of APIs that do not biotransform either sorb to MSW or enter the leachate. Sorption of APIs to MSW in a landfill is estimated using MSW generation[21] and decay[28,32] rates, leachate generation rates,[34] and one-parameter linear free energy relationships (opLFERs).[35] An opLFER linearly correlates an API's octanol/water partition coefficient with its MSW/leachate partition coefficient.[35,36] Leachate is sent to a WWTP for treatment according to current U.S. landfill practices.[28] APIs in this leachate have the same fate as APIs disposed of by the toilet disposal method.

Toilet. In the toilet disposal option, participants flush pharmaceuticals down the toilet and place associated packaging in the trash. It is assumed that each person disposes of unused APIs in a sole purpose flush, at most once a month and at least once a year. Electricity required to treat this wastewater is estimated by assuming typical energy values for domestic wastewater.[37] The values used in the model (530−1100 kWh/million gallons) include only nonaeration energy demands since the oxygen demand of the wastewater from flushing APIs is significantly lower than domestic wastewater.

Flushed APIs undergo aerobic, conventional activated sludge treatment. These APIs are removed from the wastewater's liquid phase by sorption, removed by biotransformation, or discharged with the effluent. The total mass percentage removed from the WWTP effluent, by sorption and biotransformation, is based on each representative API's observed removal during aerobic wastewater treatment (see Table 1).[38−44] The quantity of APIs sorbed is determined using typical mixed liquor concentrations[37] and opLFERs, which linearly correlate an API's octanol/water partition coefficient with its biomass/wastewater partition coefficient.[45,46] The quantity of APIs biotransformed is the difference of the total expected removal and calculated sorption. Heterotrophic yields, from zero to typical observed values,[37] are used to estimate the amount of biotransformed APIs assimilated by biomass.

The biomass formed by API assimilation and the sorbed APIs undergo solids management. It is assumed APIs remain sorbed during sludge stabilization,[3] so sorbed APIs are transported with biosolids from the WWTP to three disposal locations:[47] a landfill (13%), an incinerator (25%), or a land application site (62%). All APIs sorbed to biosolids are also expected to sorb in the landfill, which has a much higher solids concentration. The incineration of biosolids renders the sorbed APIs inert. Land-applied APIs are considered an emission to the environment. Land application sites for biosolids include agricultural lands, forests, reclaimed areas, and residential lands.[47]

Take-back. For take-back, participants drive a personal vehicle from their residence to the closest pharmacy to return unused pharmaceuticals and packaging. The nine U.S. rurality categories defined by the U.S. Office of Management and Budget[48] are used to estimate personal driving distances. Take-back participants are proportionally assigned to one of the nine rurality categories based on the geographic distribution of the U.S. population.[49] Three cities were selected to represent metropolitan counties with populations of more than 1 million (Chicago, IL), 1 million to 250 000 (Bridgeport, CT), and less than 250 000 (Bay City, MI). Six cities were selected to represent nonmetropolitan counties, both adjacent to and not adjacent to a metropolitan area, with populations of more than 20 000 (Georgetown, DE and Coquille, OR), 20 000−2 500 (Abbeville, SC and Baileyboro, TX), and less than 2 500 (Woodruff, UT and Plankinton, SD). For each of these representative cities, Google Maps was used to estimate the distances from 50 random addresses to their closest pharmacies. Additional factors considered when calculating personal driving emissions include the type of personal vehicle, the number of return trips a participant makes in one year, how many of these trips are combined with other errands, and what percentage of a combined trip's miles are allocated to taking back unused pharmaceuticals.

Once at the pharmacy, the unused pharmaceuticals and packaging are placed in a collection bin, which is a steel barrel with a removable, cardboard box liner.[9−11,50] Full cardboard boxes are transported to a warehouse for secure storage. Once enough boxes accumulate to completely fill a truck, they are hauled to the closest of the 22 commercial hazardous waste incinerators[51] for destruction. The resulting ash is hauled to the closest of the 21 commercial hazardous waste landfills in the U.S.[52]

Uncertainty and Sensitivity Analysis. A Monte Carlo analysis was undertaken to estimate the aggregate impact of parameter uncertainty on the 10 emissions categories for each disposal option. Table S7 in the Supporting Information lists the 62 uncertainty parameters, which represent energy consumption values, energy sources, API fate, API chemistry,

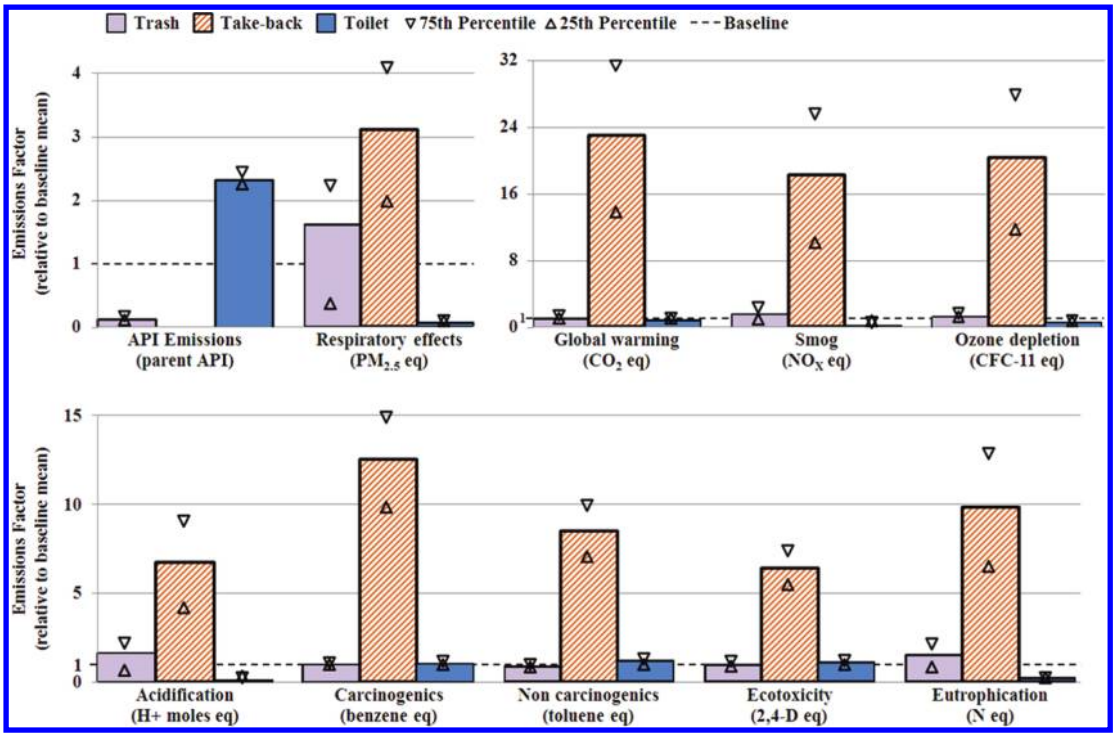

Figure 2. Results for all 10 emissions categories are presented for 100% participation in each disposal option. Bars are mean values, and triangles represent the 75th and 25th percentiles from the Monte Carlo simulations; all are relative to the baseline scenario (40% toilet, 60% trash). Table S6 lists the values for the mean and percentile (10, 25, 50, 75, and 90) values.

driving behaviors, and vehicle fuel economy. Each uncertainty parameter has a maximum and minimum value assigned from the literature or determined based on what is physically possible. Parameter ranges are characterized with a uniform distribution given the absence of data to justify assigning any other type of probability distribution (including the absence of mean values). In addition, the ranges are selected to be as broad and conservative as possible so that observed differences in emissions between disposal options are likely to be real. The uncertainty ranges for emissions associated with each disposal option are calculated from 100 000 Monte Carlo simulations. A sensitivity analysis was also conducted with the Monte Carlo simulations to determine the sensitivity of each emissions category to each uncertainty parameter. An emissions category was defined as "sensitive" to an uncertainty parameter if the resulting correlation coefficient is greater than +0.8 or less than −0.8.

Data Presentation. Emissions data for each disposal scenario are presented in decimal fraction (as emission factors) relative to the emissions of a baseline scenario reflecting current disposal practices for unused pharmaceuticals. Specifically, the baseline scenario for the U.S.[53] is taken as 40% toilet disposal and 60% trash disposal. The emission factor is greater/less than 1.0 for each emission value above/below the baseline. All results presented are the mean values from the Monte Carlo simulations and include an uncertainty range defined by the 25th and 75th percentile values.

■ RESULTS AND DISCUSSION

Figure 2 compares emissions for 100% trash, toilet, and take-back disposal compared with the baseline scenario. It is observed that implementation of take-back programs can eliminate API emissions but results in a significant increase in all non-API emissions. It is also observed that trash disposal can significantly reduce API emissions relative to the baseline without significantly increasing non-API emissions. The Monte Carlo analysis shows that these observations hold when considering the uncertainty in the model parameters.

Toilet. Figure 2 shows that flushing all unused pharmaceuticals down the toilet will more than double API emissions relative to the baseline. It also shows that emissions of carcinogens, noncarcinogens, and ecotoxins increase, though slightly. This is mostly due to the increase of biosolids incineration compared to the baseline. Because the 100% toilet scenario does not require garbage truck transport and only requires the hauling of wastewater treatment byproducts (e.g., biosolids), emissions of eutrophication, smog, and respiratory impacting substances are significantly less than the baseline. Toilet disposal requires about 15 gigawatt-hours per kilogram of unused pharmaceutical (GWh/kg API), which is 8% less energy than the baseline scenario (16 GWh/kg API). Overall, toilet disposal has the highest API emissions, the lowest non-API emissions, and the lowest energy intensity.

Trash. The fate estimates for landfilled APIs suggest that most APIs sorb to MSW and are therefore retained in a landfill. As a result, 100% trash disposal reduces API emissions relative to the baseline by 85%−92%, with a mean reduction of 88%. Given that the landfill model generally used high leachate and MSW decay rates, the 88% API reduction result is likely to be a conservative estimate for 100% trash disposal. Much higher retention of APIs in landfills is possible, especially in arid regions that have low leachate generation rates.[32]

Trash disposal slightly decreases emissions of carcinogens, noncarcinogens, and ecotoxins relative to the baseline. This is due to the decrease in WWTP biosolids production and incineration as well as the electricity offset by recovering energy from 31% of the LFG produced at the landfill. Trash disposal increases the emission of greenhouse gases and ozone depleting

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substances relative to the baseline by 10% and 30%, respectively. Also, emissions in the categories of acidification, eutrophication, respiratory effects, and smog all increase by at least 50% relative to the baseline. Most of these emissions are from the collection and incineration of MSW as well as from LFG, specifically the 69% that is flared or directly emitted to the atmosphere. The total energy required by trash disposal is 17 GWh/kg unused pharmaceutical, resulting in a 5% increase from the baseline scenario's energy intensity.

Take-back. Hazardous waste incineration and then landfilling of unused pharmaceuticals is expected to eliminate all associated API emissions to the environment. Figure 2 also indicates that all non-API emissions increase by more than 200% relative to the baseline. Emissions of carcinogens, noncarcinogens, and substances with eutrophication potential increase by more than 700%. Emissions of global warming, ozone depleting, and smog forming compounds increase by more than 1700%. The increase in global warming compounds represents 1.1−2.6 million metric tons of CO_2 equivalent emissions, which is roughly equivalent to the annual carbon dioxide emissions from a 100−300 MW coal-fired power plant.[54] Take-back disposal requires 106 GWh/kg of unused pharmaceutical, which is 560% of the energy demanded by the baseline scenario.

Further work was undertaken to understand how modifications to take-back disposal procedures affect the overall comparison among the three disposal options. One observation from the data is that most of take-back's non-API emissions arise from personal driving (it contributes more than 65% of the emissions in six non-API emissions categories). Also, 72% of the personal driving miles are in rural areas. Therefore, a "best-case" take-back disposal scenario was created that minimizes personal driving by making take-back available only in metropolitan areas (83% national take-back disposal) and assuming half of the metropolitan residents walk to a pharmacy instead of driving (41.5% national take-back disposal via walking). The unused pharmaceuticals located in rural areas are disposed of according to current disposal trends, which translates into 7% national toilet disposal and 10% national trash disposal. In this scenario, API emissions are reduced by 83% relative to the baseline. This "best-case" take-back disposal scenario has more API emissions than the 100% trash scenario (88% API emissions reduction) while still leading to significant increases in non-API emissions relative to the baseline and the 100% trash scenario. In other words, a strategic implementation of take-back programs in metropolitan areas would likely have both higher API and higher non-API emissions than 100% participation in a nationwide trash disposal program.

Participation Rates. The analyses with 100% participation show, relative to the baseline, that take-back achieves 100% reduction of API emissions at 106 GWh/kg API, and trash achieves an 88% reduction at 17 GWh/kg API. Comparing these results, the following can be asked: (1) whether the additional API reduction of 12% is worth the increase in non-API emissions and financial costs of implementing take-back programs and (2) whether this difference in API emissions is maintained when considering foreseeable participation rates for each disposal option.

With respect to the second question, two U.S.-based surveys suggested that 74%-90% of respondents would likely participate in a take-back program.[9,15] However, a pilot take-back program in Clark County, Washington yielded a participation rate of less than 0.1%.[11] Similarly, after 40 years of a national take-back effort in Sweden, the participation rate has been estimated at only 43%.[16] In addition, it has been reported that more than 50% of people in Sweden store unused pharmaceuticals at home for prolonged periods,[16] which runs counter to the ONDCP goal of safe and fast disposal of unused pharmaceuticals.

To understand the effect of foreseeable participation rates on emissions, a disposal scenario was created where 43% of U.S. households participate in take-back programs and the remaining 57% use trash disposal. Under this scenario, analogous to Swedish participation rates, API emissions are reduced by 93% while all non-API emissions increase by 130−950% relative to the baseline. Under the unlikely case where 43% of unused pharmaceuticals are taken back without any personal driving, the 93% reduction in baseline API emissions still increases all non-API emissions by 30−270% relative to the baseline. Figure 3 summarizes these results and shows, using

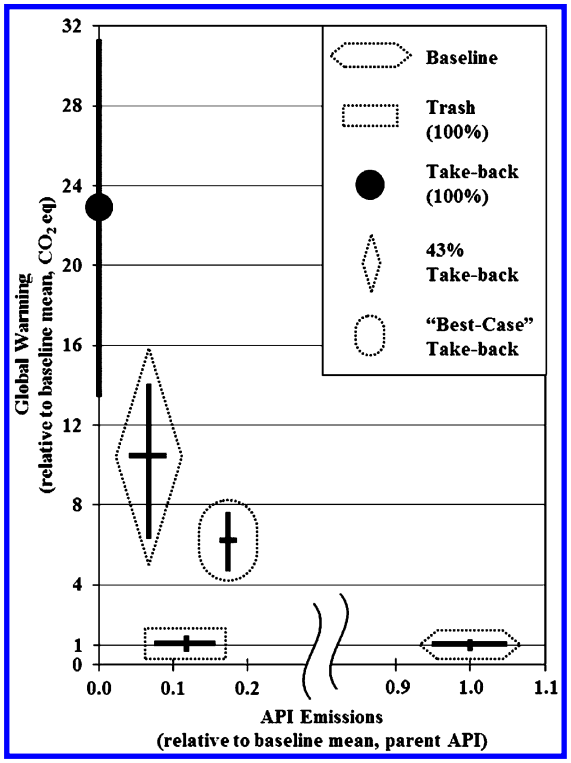

Figure 3. Results are presented for each scenario's global warming emissions as a function of their API emissions. The uncertainty lines (dark solid lines) show the range of values between the 25th and 75th percentiles. Shapes designate the following disposal scenarios: baseline (40% toilet, 60% trash); 100% trash; 100% take-back; 43% take-back (43% take-back, 57% trash); and "best-case" take-back (take-back only in metropolitan area with half not driving, 10% trash, 7% toilet).

global warming emissions as an illustrative example, that a take-back program achieving greater reductions of API emissions than a 100% trash disposal scenario will have higher non-API emissions.

Disposal Recommendations. If the contribution of environmental API emissions from the disposal of unused pharmaceuticals is determined to be negligible relative to the API contributions from human and animal excretion, then toilet disposal would be the best approach. Toilet disposal has the fewest non-API emissions and is the easiest and fastest way for individuals to remove pharmaceuticals from their homes. Toilet

disposal would also be viable if technology could be easily and inexpensively added to all WWTPs to render APIs and their transformation products harmless.

If it is decided that all sources of environmental API emissions should be reduced immediately, then take-back or trash disposal is necessary. Under a take-back participation rate of 43%, take-back reduces 93% of API emissions relative to the baseline (assuming the other 57% is trashed). Trashing all unused pharmaceuticals can reduce baseline API emissions by 88%. Although the implementation of take-back programs might achieve a 5% improvement in API reduction compared to trash disposal under Swedish participation rates, it would come with significant downsides: (1) non-API emissions would increase significantly, (2) societal costs would increase significantly (estimated at 2 billion dollars per year for a nationwide program[14]), (3) disposal inconvenience would increase significantly,[10,15] and (4) home storage of unused pharmaceuticals may increase to an unacceptable level (as in Sweden) due to disposal inconvenience, which increases the risk of poisoning, abuse, and addiction.[18]

Taken together, the results indicate that trash disposal would be nearly as effective in reducing environmental API emissions as take-back programs but without significant increases in non-API emissions or societal costs. Furthermore, since 60% of individuals in the U.S. already trash their unused pharmaceuticals, trash disposal is likely to accomplish faster removal of unused pharmaceuticals from households due to higher participation rates and greater convenience.

A re-evaluation of disposal options for unused pharmaceuticals may become necessary as future waste management strategies, technologies, and research improve our understanding of environmental API sources and impacts. In the meantime, the management of unused pharmaceuticals with MSW can provide a disposal option that is likely to have a high level of compliance, lower costs, and ultimately a similar degree of API removal as compared with take-back programs.

■ ASSOCIATED CONTENT

Supporting Information

More information about data, assumptions, calculations, and uncertainty parameters. 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.

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