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Pharmaceutical compound content of municipal solid waste

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ABSTRACT

The occurrence and fate of pharmaceuticals in landfills has been largely neglected. Once discarded in municipal solid waste (MSW), pharmaceuticals within a landfill may undergo degradation, adsorption, or enter the leachate and eventually exit the landfill. The active pharmaceutical ingredient (API) concentration of MSW was predicted using available statistics on medication usage and directly measured by a MSW composition study. Estimation calculations resulted in a potential concentration of APIs from 7.4 to 45 mg/kg of MSW, varying with the percentage of dispensed medications assumed to become unused. Direct measurement resulted in the collection of 22 APIs comprising a total of 22,910 mg. This resulted in a final concentration of 8.1 mg/kg within MSW. Additionally, 45 empty medication containers were collected which potentially contained 33 differing APIs upon disposal.

1. Introduction

Recent studies have shown pharmaceutical compounds entering sewage treatment systems are not fully removed and are discharged to the aquatic environment [1–5]. In 2002, the United States Geological Survey released a national reconnaissance of pharmaceuticals, hormones, and other organic pollutants in surface waters that revealed the presence of these compounds in more than 80% of the streams tested [6]. The release of these compounds to the environment has raised concerns over their effects as studies have emerged showing their potential influence on wildlife and possible changes upon biological systems [7–11].

Previously, the recommended disposal method of unused pharmaceuticals was the sewage system. This was chosen as a means to protect children from accidental poisoning in the home, prevent animal poisoning from scavenging either at the home or the landfill, and more recently to prevent scavenging of prescription medications for illicit use. However, despite this recommendation the use of municipal refuse and landfills for pharmaceutical disposal was not uncommon. In a recent pharmaceutical collection project, patrons were surveyed to determine the methods of waste disposal they used prior to the collection program. Nearly half of the survey participants indicated that they had used their household trash as a disposal method, just slightly less than those who had used sewer disposal [12]. Only a limited number of studies have investigated the presence of pharmaceutical compounds in landfills. These studies have focused on the analysis of landfill leachate [13–15] or leachate-contaminated groundwater [14–19]. Even fewer studies have examined the potential pharmaceutical waste composition of municipal solid waste (MSW) [20,21].

Few specific regulations apply to the management of discarded pharmaceuticals, particularly household medications, and myriad of pathways for disposal may be followed [22]. Due to emerging research highlighting the presence and effects of pharmaceuticals in aquatic environments, many U.S. states have begun to recommend refuse disposal instead of sewage disposal to their residents [23–26]. In February 2007, the United States White House Office of National Drug Control Policy released guidance on the proper disposal of unused or unwanted prescription drugs [27]. The guidance directs consumers to dispose of the unused drugs in household trash or to take advantage of drug take-back programs, rather than flushing the drugs down the toilet. These new policies will undoubtedly result in increased amounts of pharmaceuticals entering MSW landfills.

The occurrence and fate of pharmaceuticals in landfills has been largely neglected. Once discarded in MSW, pharmaceuticals may undergo degradation, adsorption, or enter the leachate percolating through the landfill. In modern lined landfills, leachate is collected and often treated using sewage treatment works, the very system the new policies were designed to avoid. While studies have indicated landfills as potential sources of APIs to the environment, no research could be located to quantify the input of APIs to landfills and how the quantity of APIs entering MSW affects their fate and potential leachate concentrations. The objective of this study was to determine the current API concentration of MSW using two

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methods: (1) mathematical estimation based upon statistics of usage and consumer disposal methods and (2) direct MSW composition measurement. These results may then form the basis in formulating strategies for future pharmaceutical waste management and research concerning pharmaceutical emission from landfills.

2. Methods

2.1. Mathematical estimation

Information concerning the total mass of a pharmaceutical distributed is not widely published, with the total dollar value being much more common. However, in Australia, the Drug Utilization Sub-Committee (DUSC), formed in 1988, annually assembles data on prescription drug usage. The DUSC publishes the World Health Organization's defined daily dose (DDD) for each prescription drug measured in units of milligrams per 1000 head of population, per day. In a previous study, Fisher and Borland [28] used this report to estimate the pharmaceutical burden to the environment in Sydney. Employing a similar technique, it was possible to estimate the input of pharmaceuticals to landfills in the United States.

Fig. 1 illustrates the method and statistical values utilized. The United States and Australia are similar in health care development and medication availability [29]. Additionally, the types of medication used as similar as seen by the top 10 prescribed medications in Australia in 2004 being among the top 25 medications used in the United States [30,31]. Therefore, the per capita medication usage of Australia may be converted to a corresponding per capita usage of the United States. This was accomplished utilizing a ratio of the prescriptions per person in the United States versus Australia [32–34]. Using the U.S. population, this value was then equated to the total annual active pharmaceutical ingredient (API) usage in the United States. The annual total API usage was then corrected based upon published research and governmental statistics. These factors included: (1) over-the-counter (OTC) pharmaceutical usage [35,36], (2) the percentage of medications which go unused [36–39], and (3) percentage of unused medications discarded to MSW [12]. Based upon these values and the total annual MSW disposal rate of the United States, the concentration of API within United States MSW was predicted.

2.2. Direct measurement

A waste composition study of MSW was conducted to measure the percentage by mass and types of pharmaceuticals in the MSW stream of Orange County, Florida. The study was conducted in December 2006 at a MSW transfer station. The choice of a transfer station afforded four primary advantages: (1) a covered location to reduce the impact of inclement weather, (2) reduced potential error introduced from landfill soil becoming intermingled with selected trash, (3) ease of obtaining samples due to the immediate access to equipment at the facility, and (4) a concrete partition within the facility to provide additional safety for personnel from operating machinery and waste vehicles.

2.2.1. Waste sectors

The composition of waste from residential and commercial sectors was independently determined during the study. Residential wastes were comprised of MSW from single-family and multi-family residential dwellings. Commercial sectors included commercial and institutional businesses of any type, including offices, restaurants, retail establishments, warehouses, hotels, schools, and government buildings. It is noted that samples were not taken from segregated loads of bulky items, recovered materials, or solely construction and demolition debris. Furthermore, commercial waste from the medical industry, such as doctor offices, nursing homes, and hospitals was not targeted or known to be included within the waste examined.

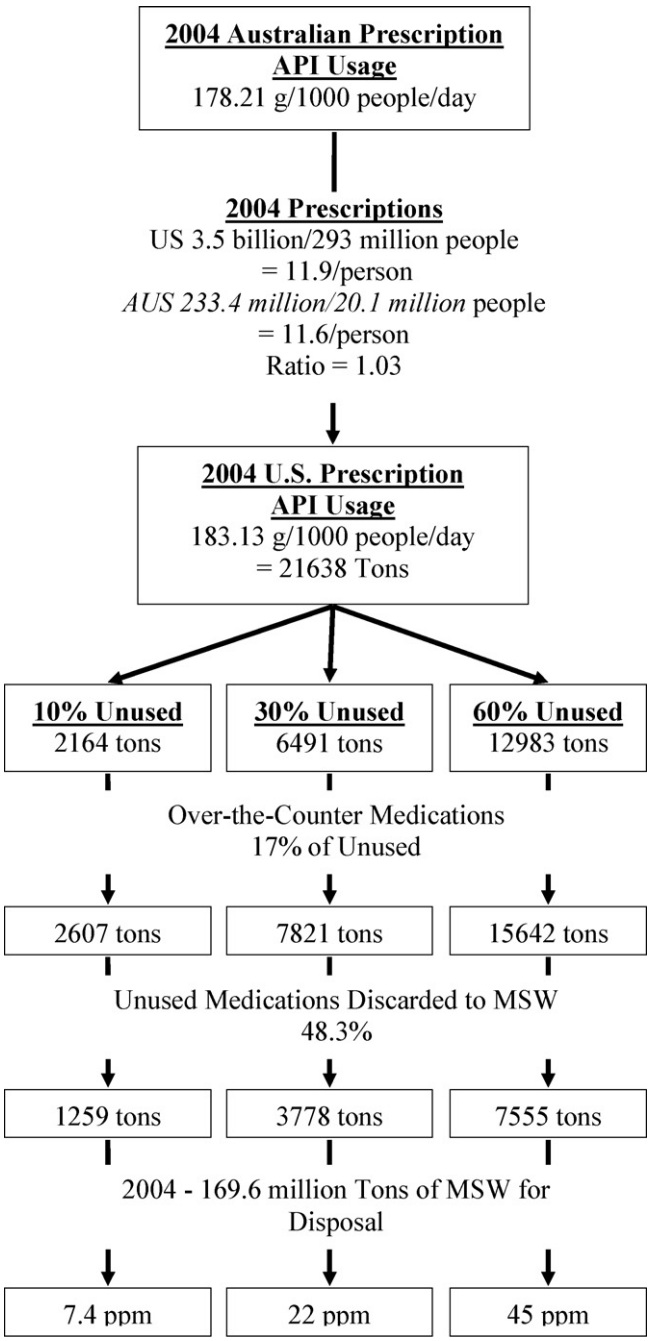

Fig. 1. Prediction of United States municipal solid waste active pharmaceutical ingredient concentration.

2.2.2. Sampling and sorting

Sampling and sorting was conducted using standard industry practice [40,41]. The commercial or residential origin of each MSW sample was verified by the transfer facility staff through interview with the collection vehicle driver. Once identified, a "grab" sample of MSW, visually estimated to weigh from 200 to 400 pounds, was obtained from the MSW on the tipping floor of the facility. Industry

standard practice specifies a minimum average sample size of 200 pounds. However, variation in load materials and their density can result in some samples weighing less or more than the 200-pound target. Due to this, efforts were made to ensure that samples were of significant size to achieve well greater than 200 lbs on average. To minimize bias, a vertical slice was taken from the waste stack and the sample was compared to the visible characteristics of the full waste load for any obviously non-representative material prior to sorting.

The MSW was segregated into 10 major solid waste categories including paper, plastic, glass, iron, aluminum, food, yard waste, and textiles. Full sorting of the selected MSW permitted comparison to US national waste composition data to ensure the waste selected was typical of the national average. Each sample was manually loaded onto a sorting table with bagged waste carried to the table and loose waste transferred via plastic containers. Large or bulky items, such as tires, were separated and weighed directly. The sorting table was covered by 1/2-in. (1.2 cm) screening and particles small enough to fall through the screen were examined for loose medication capsules or tablets and then characterized as organic fines. Remaining waste was manually sorted into bins labeled for each group with a separate bin specific for medication containers and loose medications. Bagged and boxed materials were opened and all waste sorted. Upon completion of sorting each sample load, each group was weighed to determine its individual weight. Pharmaceutical wastes were then further processed to determine the quantity (number of tablets or volume of liquid), API, and medication brand information.

3. Results

3.1. Statistical estimation

In Australia, approximately 233.4 million prescriptions were filled in 2004 [34]. With a population at that time of 20.1 million people, a total of 11.6 prescriptions per person were filled. In 2004, the number of retail prescriptions filled in the United States was 3.27 billion [33] and the number of outpatient prescriptions filled by veterinary facilities, hospitals, and other medical clinics was estimated to be 229.9 million [32], for a total of 3.50 billion prescriptions. Dividing this by the United States population for 2004 (293,638,158), the average number of prescriptions per person in the U.S. was 11.9 or a factor of 1.03 greater than in Australia.

The average consumption of prescription pharmaceuticals per thousand people in Australia was calculated to be 178.21 g of APIs per day in 2004 [34]. Thus, as shown in Fig. 1, multiplying the Australian consumption by the 1.03 factor yields a total of 183 g/day per thousand people in the United States. It should be noted that this conversion relies upon the types of medications used within the United States and Australia being similar. This was supported by the prior comparison of the top prescribed medications of each country and the similar prescription rates. Differences in these quantities would be a source of error in the estimation. In the United States, 183 g/day per thousand people is equivalent to a total of 2.16×10^4 tonnes/year of active ingredients purchased via prescription.

To determine the amount of the dispensed APIs entering MSW, the percentage of the prescribed drugs which become unused is required. The measure of prescription fulfillment by a patient is termed “compliance” or “concordance” by the medical community and includes factors such as consuming all medication prescribed, taking the medication at the appropriate time and under the appropriate conditions (such as avoiding confounding factors that may reduce its effectiveness). Studies of prescription compliance can be found extensively in the literature, but specific details on the remaining unused medication quantity are seldom mentioned. In a study conducted in Alberta, Canada, it was estimated that 60% of the original drugs dispensed in a prescription were returned to the local pharmaceutical collection program [38]. In another German study, it was determined that 65% of the original medication was returned [37]. Other studies have measured only 3% [36] or estimated 4–15% [39]. In light of the wide range of potential values, three percentages were used to estimate the amount of medication which becomes unused—10, 30, and 60%. These values (10 and 60%)were chosen to approximate the extreme upper/lower values and an average value (30%) of the prior studies.

To this point, only prescription medications have been included in the estimation. Nonprescription or OTC medications may also be discarded and must be included in the estimation. Garey et al. [35] reported 65% of medications collected in a Houston, Texas program were prescription drugs, while 27% were OTC and 8% were samples. A Swedish collection program found that only 7% of the unused medications were OTC drugs [36]. An average of these two studies, 17%, was assumed as the percentage of unused medications attributable to OTC medications.

The disposal options for unused medications practiced by consumers consist primarily of flushing to the sewage system, household trash, and return to a pharmaceutical collection program. The result of several studies on pharmaceutical disposal trends has been previously published. Musson et al. [12] cited four studies which measured the disposal options practiced by consumers. The percentage of people utilizing MSW disposal averaged 48.3% over these four studies and the total APIs for disposal were multiplied by this percentage. As seen in Fig. 1, the total APIs entering US landfills in 2004 are estimated to be 1.26×10^3 tonnes (10% unused), 3.78×10^3 tonnes (30%), and 7.56×10^3 tonnes (60%).

These computations account for only the active ingredients of the medications and do not account for the salts and other materials included in the formulations which comprise a much larger percentage of the medicine. Dividing the total APIs landfilled by the total of 169.6 million tonnes of MSW landfilled in 2004 [42], the average concentration of pharmaceutical compounds in waste deposited to US landfills in 2004 was 7.4 mg/kg (10%), 22 mg/kg (30%), or 45 mg/kg (60%). These concentrations encompass all pharmaceuticals and the concentration of any single pharmaceutical can be expected to be much lower.

3.2. Direct Measurement

A total of 6204 lbs (2820 kg) of MSW was sorted in 22 samples comprised of an equal number of residential and commercial loads. Samples ranged in weight from 163.9 to 437.9 lbs, with an average of 282.0 ± 68.9 lbs. The composition of the waste was verified to be typical of United States MSW as determined by comparison of the percentage by weight of each of the 10 waste categories with national averages. The pharmaceutical compounds collected during the composition study are shown in Table 1. A total of 22 differing APIs were collected comprising a total of 22,910 mg. This resulted in a final concentration of 8.1 mg/kg within the MSW, a value of the same magnitude predicted in the prior calculations.

Of the MSW received from commercial sources, only 1 out of 11 samples contained a measurable API, ciprofloxacin. The remaining APIs collected during the study were received from 9 out of the 11 residential MSW samples studied. However, in both sources a large number of empty pharmaceutical containers were located. It was not possible to determine if these were discarded as empty containers or became ruptured during collection and transport and their contents lost, dissolved, or crushed within the MSW. As an example, two of the collected APIs, principen and lexapro, were

Table 1
APIs measured in the MSW of Orange County, FL

API[a]	Pharmaceutical category	Form of delivery (liquid/solid)	Number of containers recovered	Total quantity (mg)
Ciprofloxacin HCl	Antibiotic	Solid	1	6500
Acetaminophen	NSAID	Liquid/solid	3/1	5380
Amoxicillin	Antibiotic	Liquid	1	4000
Ampicillin	Antibiotic	Solid	1	3000
Donpezil HCl	Alzheimer's disease	Solid	1	1050
Ibuprofen	NSAID	Liquid	1	900
Pseudoephedrine HCl	Decongestant	Liquid/solid	3/1	737
Metoprolol succinate	Antihypertensive/beta blocker	Solid	1	475
Bismuth subsalicylate	Gastrointestinal	Liquid	1	349
Dextromethorphan HBr	Cough suppressant	Liquid/solid	3/1	306
Minoxidil	Hair loss	Liquid	1	100
Albuterol sulfate	Bronchodilators	Liquid	3	26.4
Tegaserod maleate	Irritable bowel syndrome	Solid	1	24
Cetirizine HCl	Antihistamine	Solid	1	20
Polymyxin B sulfate	Antibiotic	Liquid	1	16.7
Trimethoprim	Antibiotic	Liquid		10
Clobetasol propionate	Corticosteroid	Liquid	1	5.7
Phenylephrine	Decongestant	Solid	1	5
Brompheniramine maleate	Antihistamine	Solid	1	2
Nicotine	Smoking treatment	Solid	1	2
Levothyroxine sodium	Hypothyroidism	Solid	1	0.75
Clotrimazole	Antifungal	Solid	1	0.3

[a] Some medications, such as cold medications, contained more than one API.

collected as loose tablets and capsules within the waste and were identified through medication markings. Therefore, the potential for additional medications not within a container or separated from their container to become crushed or dissolved exists and the actual API concentration to be greater than measured. The 33 APIs listed on the 45 empty containers is given in Table 2.

4. Discussion

In a recent pharmaceutical industry report on the potential releases of medicines in MSW landfill leachate, calculations were based upon the assumption of 5, 10, and 15% of all sold medications were disposed via landfills [20]. This would be equivalent to 10–30%

Table 2
APIs of empty medication containers within the MSW of Orange County, FL

API[a]	Pharmaceutical category	Form of delivery (solid/liquid)	Number of containers recovered
Acetaminophen	NSAID	Solid	6
Amlodipine besylate	Hypertensive	Solid	1
Aripiprazole	Schizophrenia/mania	Solid	1
Atorvastatin calcium	Cholesterol	Solid	2
Azithromycin	Antibiotic	Solid	1
Caffeine	Stimulant	Solid	2
Celebrex	NSAID	Solid	1
Chlorpheniramine	Antihistamine	Solid	1
Dexamethasone	Corticosteroid	Liquid	1
Dextromethorphan	Cough suppressant	Solid	1
Diazepam	Antianxiety	Liquid	1
Diclofenac	NSAID	Solid	1
Digoxin	Heart failure	Solid	1
Dutasteride	Prostate enlargement	Solid	1
Esomeprazole	Gastrointestinal	Solid	7
Ethinylestradiol	Oral contraceptive	Solid	2
Genotropin	Growth hormone	Solid	1
Glycerine	Laxative	Liquid	1
Ibuprofen	NSAID	Solid	3
Losartan potassium	Antihypertensive	Solid	1
Methscopolamine	Anticholinergic	Solid	1
Metoprolol succinate	Antihypertensive/beta blocker	Solid	1
Montelukast	Asthma/allergy	Solid	1
Neomycin	Antibiotic	Liquid	1
Norethindrone	Oral contraceptive	Solid	2
Pamabrom	Diuretic	Solid	1
Pseudoephedrine	Decongestant	Solid	1
Pyrilamine maleate	Antihistamine/diuretic	Solid	1
Quinapril	ACE inhibitor	Solid	1
Simethicone	Gastrointestinal	Solid	1
Thiabendazole	Antifungal	Liquid	1
Tussin ex	Expectorant	Liquid	1
Ziprasidone HCl	Schizophrenia/mania	Solid	1

[a] Some medications, such as cold medications, contained more than one API.

Table 3
Comparison of Orange County, Florida and United States National Demographic Data[a]

Demographic category	Orange County, FL	United States
Persons under 5 years old, 2006	7.5%	6.8%
Persons under 18 years old, 2006	25.3%	24.6%
Persons 65 years old and over, 2006	9.6%	12.4%
Persons per household, 2000	2.61	2.59
Median household income, 2004	$41,725	$44,334
Persons below poverty, 2004	12.6%	12.7%
Healthcare, percent uninsured, 2000	17.3%	14.2%

[a] *Source*: U.S. Census Bureau Quickfacts.

of all medications becoming unused if it is assumed that approximately 50% of unused medications are discarded in the trash [12]. Thus the mathematical estimation based upon 10, 30, and 60% of medications becoming unused is reasonable when compared to this study.

The large range in statistically estimated API concentration in MSW, 7.4–45 mg/kg is due to the large uncertainty in the quantity of medications which become unused once given to the patient/consumer. The direct measurement of 8.1 mg/kg API concentration is equivalent to a calculated value of 11% of all medications becoming unused. However, the percentage of medications which become unused may be lower as the prior calculations were unable to account for internet prescription sales, an increasingly popular source of prescription medications. This would result in a higher initial API input. Additionally, an increase in number of prescriptions between the calculated year (2004) and the measurement year (2006) may result in an increase in the measured API concentration over the predicted concentration.

The pharmaceuticals comprising the greatest API concentration in MSW included antibiotics (ciprofloxacin and amoxicillin) and nonsteroidal anti-inflammatory drugs (acetaminophen and ibuprofen). The Alzheimer's treatment, ampicillin, which was collected from a single residential sample, also appeared in significant quantity but may be skewed due to this one-time disposal. The large quantity of cold medications including antihistamines, decongestants, cough suppressants, and fever reducers may be due to the time of year in which the study was conducted and other medications may be expected to be prevalent during other seasons, such as allergy medications in the spring.

Additionally, population demographics may also affect the expected results. A comparison of relevant demographic statistics of the test location with the United States National averages is shown in Table 3. As seen in this table, the demographics of the test location do not differ largely from the national values. The lower percentage of residents above the age of 65 may result in the API concentration being lower than the national average as people of this age group have been shown to use medications more often [12,43]. Additionally, the greater percentage of residents without health insurance may impact the availability of medications and result in a lower API concentration at the test site.

The concentration determined in this study may underestimate the final disposal of pharmaceutical compounds within a landfill. Beyond the direct disposal of unwanted pharmaceuticals, pharmaceutical manufacturing wastes in landfills and sewage treatment sludge sent to landfills are also a potential source of pharmaceuticals. Pharmaceuticals adsorbed to sewage solids may become released within the landfill environment and enter the landfill's leachate [1,44]. Furthermore, new government policies to direct unused medications to MSW disposal may result in a significant increase, possibly doubling the measured concentration if the roughly 50% of discarded medications previously flushed to the sewage system become discarded within MSW.

This research did not attempt to correlate the compounds measured within the MSW to their concentration within landfill leachate nor determine their potential for release from a landfill. However, this study does address a current gap of knowledge in the lifecycle of pharmaceutical compounds. Although, a large and growing number of research studies have examined the occurrence and fate of pharmaceuticals in municipal wastewater treatment, few have addressed their presence in landfills and even fewer have attempted to quantify their concentrations. Using this data and prescription or distribution rates for individual compounds, researchers and regulators may be able to more accurately predict the landfill concentrations expected for individual compounds using the presented mathematical method and direct measurement data.

5. Conclusions

The API concentration of MSW was mathematically estimated using existing data on pharmaceutical use and disposal to be between 7.4 and 45 mg/kg. This concentration varied with the percentage of dispensed medications assumed to become unused. Direct measurement resulted in the collection of 22 APIs comprising a total of 22,910 mg from approximately 2800 kg of MSW. This resulted in a final concentration of 8.1 mg/kg within MSW. This corresponded with an estimation of 11% of all medications becoming unused. Additionally, 45 empty medication containers were collected which potentially contained 33 differing APIs upon disposal. Thus, the reported 8.1 mg/kg is the minimum measured API concentration and the actual concentration may be greater due to input not measured from the empty containers. This research provides information pertinent to the life-cycle analysis of pharmaceutical compounds and when coupled with landfill leachate data enables the assessment of the effectiveness of landfill disposal of pharmaceutical compounds.

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