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Material Flow Analysis of Carbon Nanotube Lithium-Ion Batteries Used in Portable Computers

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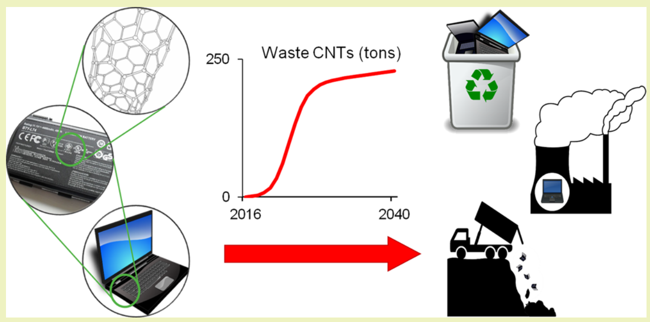
Supporting Information

ABSTRACT: Engineered nanomaterials are finding application in a wide range of consumer electronics. In particular, carbon nanotubes (CNTs) are candidate materials for use in enhancing the performance of lithium-ion battery anode and cathodes. However, past studies indicate that some toxicological effects exist for CNTs, although full evaluation may yet take time. Appraisals of material flows of potential products containing CNTs are useful for early recognition of environmental problems, for investment planning in production and waste management infrastructures, and for government policy formulation. This material flow analysis (MFA) study uses a stock dynamics and logistic model to forecast the technology transition from conventional Li-ion batteries in portable computers to CNT Li-ion batteries and the subsequent waste generation of CNTs in obsolete laptop batteries. State-specific recycling rates for electronic waste are projected to determine the quantities of CNTs in laptop batteries destined for recycling, incineration, or landfilling. As markets for CNT-enabled electronics begin to expand, United States collection and recycling facilities may consider establishment of new processes or controls to reduce the potential for CNT emissions and exposures.



KEYWORDS: *Material flow analysis, Technology substitution, Electronic waste, Lithium-ion batteries, Carbon nanotubes*

INTRODUCTION

Rapid development in technology increases the rate of electronics development and in turn the rate of waste generation caused by product obsolescence. With each new hardware release, new materials and technologies are utilized to improve product capabilities. Emerging technologies can quickly begin to outpace the rate at which goods can be evaluated for their potential environmental impacts at end-of-life (EoL).[1] Engineered nanomaterials constitute an emerging class of materials that have found application in consumer electronics,[2] including portable and desktop computers, cell phones, and batteries. Carbon nanotubes (CNTs), explored for applications primarily in consumer electronics and the energy sector, are candidate materials for use in lithium-ion batteries due to their favorable electrochemical and mechanical properties. Their excellent conductivity allows for use in a variety of modes, such as in individual nanotube electrodes, as a layer on top of an existing electrode, or as a free-standing electrode without any additional support.[3,4]

Battery anodes and cathodes made from multi-walled carbon nanotubes (MWCNTs) are being developed for commercialization and show promise for increased current capacity and battery lifetime. In the coming years, nano-enabled Li-ion batteries may replace conventional Li-ion batteries, not only as traction batteries in electric and hybrid vehicles, but also in smaller portable electronic devices such as power tools and laptop computers. Use of engineered nanomaterials other than CNTs in both positive and negative electrodes of Li-ion batteries has also been reported.[5] Nanoparticles of transition metals are often used in Li-ion battery anodes.[6] Anodes of Co_3O_4,[7,8] Mn_3O_4,[9] SnO_2,[10] and silicon-based[11] nanoparticles have displayed large specific capacities and good rate capabilities and cycling performance. Nanoparticles of $LiFePo_4$, $LiCoO_2$, and V_2O_5 are commonly used in cathode materials as well.[5]

As the number of nano-enabled products rises, so does the generation of nanomaterial-containing waste and subsequent treatment in municipal solid waste (MSW) incinerators and disposal in landfills when products are discarded.[2] Currently,

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there are no regulations in the United State specifically governing treatment or disposal of nano-enabled products. NIOSH had previously proposed a recommended exposure limit (REL) of 7 $\mu g/m^3$ in 2010, which has been reduced to a recommended working lifetime exposure limit to CNTs of 1 $\mu g/m^3$, which is the 8 h time-weighted average for 45 years.[12] This REL is based on evidence of CNT uptake and toxicity in laboratory animal studies,[13] including the MWCNTs that are planned in battery applications.

Although no direct measurement or inventory of nanomaterial disposal has been documented in the literature, previous studies have modeled projected quantities of these materials that are currently managed in MSW systems.[14−16] Looking to the future, appraisals of expected nano-containing waste flows are useful for early recognition of environmental problems, for investment planning in production and waste management infrastructures, and for government policy formulation such as environmental policy, R&D funding emphasis, or strategic material objectives.

Here, a detailed model is presented that forecasts the increase in CNT use in the specific application of Li-ion batteries in portable computers and their subsequent management as obsolete electronic waste over the next 25 years. The material flow model uses logistic functions, commonly employed to model technology transitions in previous studies,[17−19] to model the growth in portable computer sales with population and affluence; the introduction, growth, and potential saturation of CNT-enabled batteries for this application, and the generation of CNT-laden electronic waste and disposition by state. Several growth scenarios are modeled, placing useful bounds on the results.

METHODS

Material Flow Analysis. Material flow analysis (MFA) is a method used to describe, investigate, and evaluate the flows and accumulations of materials and substances through both the economy and the environment. MFA is based on a stock and flow model (principle of mass conservation), in which time step changes in stock are determined by tracking additions (flows in) and subtractions (flows out) to stock. The goal of a material flow analysis is to increase the understanding of a system defined by spatial and temporal boundaries, which is a prerequisite for better control and management.[20,21]

The relationship between stocks and flows is represented as

$$S_i = S_{i-1} + I_i - O_i \tag{1}$$

where S_i is the stock of material, or product in use in year i, I_i is the input (or sales) and O_i is the outflow of disposed products.

The generation of obsolete products O_i and sales I_i are related by the lifespan distribution L_j, which is the probability after j years that a new product becomes obsolete, as shown in

$$O_i = \sum I_{i-j} \times L_j \tag{2}$$

Consideration of Material Intensity and Stock Dynamics. This basic MFA model requires that sales data I_i be known for every year, which is not possible when forecasting. Müller et al.[22] used a stock dynamics approach as an alternative method to forecast resource demand and waste generation, in which sales become endogenous. This approach is based on physical accounting and has become common in MFA and policy analysis[23] and is used here. The central driving forces are the population and its lifestyle, which are manifested in service providing stocks of products in use. The system involves three dynamic variables: population within the region of interest (p), service units in use (s), and their associated material stocks in use (m). Each variable has a stock S, input I, and output O flow ($I^{(p)}$, $O^{(p)}$, $I^{(s)}$, $O^{(s)}$, $I^{(m)}$, $O^{(m)}$) and are related by three determinants: penetration rate N (or service units per capita, with a maximum value of $K_C^{(S)}$), materials intensity per service unit ($M_S^{(m)}$), and lifetime (L), all of which can vary through time. The overall stock of service units in use is driven by population and penetration rate. The demand for service units in use determines, in conjunction with the lifetime, how many units need to be added and how many units become obsolete. The input of new service units determines, depending on the technology, how much material is needed. This material input is used, together with the lifetime, to calculate the material stock accumulation and output of obsolete products and materials.

Logistic Model. The logistic model has its roots in ecology in modeling population growth, but it can be also applied to consumer products to solve for the penetration rate. The differential equation that describes this model is as follows

$$\frac{dN}{di} = rN\left(1 - \frac{N}{K}\right) \tag{3}$$

where N represents the penetration rate of the product, r is the intrinsic growth rate, and K represents the carrying capacity or demand saturation point, which is the maximum average number of service units per person, corresponding to $K_C^{(S)}$ in the previous section. Here, K is assumed constant, while N varies over time to produce a sigmoid or S-shaped curve. The solution of eq 3 in year i is

$$N_i = \frac{K}{e^{-(ri+C)} + 1} \tag{4}$$

where $C = ln[N_0/K - N_0]$, and N_0 is the penetration rate for the reference year or year 0. The penetration rate can also be estimated empirically in terms of the stocks of service units S_i and the population as follows

$$N_i = \frac{S_i}{Q_i} \tag{5}$$

Technology Substitution. Variations of the logistic model have been employed for decades to project transition trends for many technology systems.[24,25] The function depicted in eq 4 can be utilized to project baseline scenarios for technology change and replacements for competing electronic products.[1] Technology transitions in desktop and portable computer cases have been modeled by Bader et al.[26] as follows

$$f_n(t) = \frac{f_{s,n} - f_{i,n}}{1 + e^{-r_n(t-t_{n,0})}} + f_{i,n} \tag{6}$$

In this model, the growth rate, or market fraction $f_n(t)$ is fit for technology n, where $f_{s,n}$ is the saturation value, $f_{i,n}$ is the initial fractional value, r_n is proportional to the maximum growth or decay rate, and $t_{n,0}$ is the inflection point on the logistic curve. According to Fisher and Pry,[27] who successfully extended the model to portray market share trends for consumer goods, technology can in theory reach 100% of market share. With this consideration, eq 6 becomes

$$F_n(t) = \frac{1}{1 + e^{r_n(t-t_{n,0})}} \tag{7}$$

Forecasting Obsolete Products. Combining stock dynamics and logistic models, the forecast for a quantity of portable computers that will become obsolete and discarded is determined. The technology substitution model is used to determine the fraction of these computers that would have a nano-enabled Li-ion battery. Modifying and adding the component to eq 2 results in the following relationship for the generation of obsolete CNT-containing batteries

$$O_i^{(s)} = (\sum I_{i-j} \times L_j) \times F_j \tag{8}$$

where F_j is the fraction of portable computer Li-ion batteries that contain CNTs in year j from eq 7.

Input (sales) of laptops I_i is determined by multiplying the population stock $S_i^{(p)}$ by the penetration rate N_i in year i. Historical and

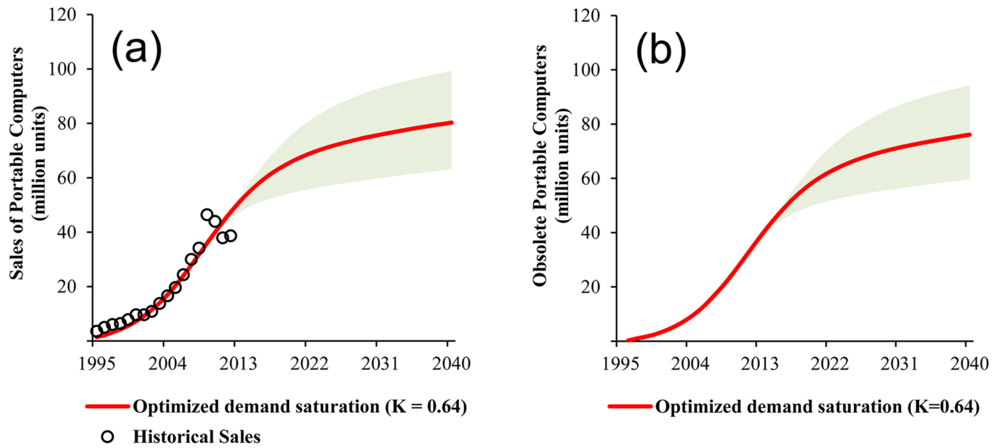

Figure 1. Forecasting (a) sales of portable computers and (b) generation of obsolete portable computers, excluding exports.

projected population data were provided by the U.S. Census Bureau.[28] The product lifetime L_j is defined as the time between product purchase in year j and when it becomes obsolete or not considered part of in-use stocks. Various statistical distributions are possible to describe product lifetimes. Yang et al.[29] considered a baseline discrete probability distribution for computer lifetimes with an average of 3 years for portable computers as shown by $P(L = 2) = 0.25$, $P(L = 3) = 0.5$, and $P(L = 4) = 0.25$. This assumption was based on previous work by Williams[30] and is consistent with available data for personal computers (desktop and laptop).[31]

Some used portable computers are exported from the United States each year. Therefore, the number of exported used laptops E should be considered in the model. Equation 2 becomes

$$O_i^{(s)} = [(\sum I_{i-j} \times L_j) - E_i] \times F_j \tag{9}$$

E is estimated following Duan et al.,[32] who calculates the number of used portable computers exported from United States by using various methods. It is estimated that on average 871,000 used laptops were exported from United States in 2010 (United States export neighborhood valley-emphasis method). The percentage of exported used portable computers in 2010 is calculated by dividing the number of exported used laptops by the total estimated number of obsolete portable computers. It is assumed that this percentage remains the same over the entire study period.

The material intensity variable $M_S^{(m)}$ from the stock dynamics approach is then used to determine the amount of CNTs inside the obsolete batteries

$$O_i^{(m)} = M_s^{(m)} \times O_i^{(s)} \tag{10}$$

Current CNT-enabled Li-ion battery designs specify 3−5 cells, with each cell containing an average of 1 g MWCNTs.[33] By fixing this CNT concentration over the entire scenario period and multiplying by the total number of obsolete units, the mass of CNTs in obsolete batteries are estimated.

End-of-Life Management. Electronic waste management in the United States is regulated primarily at the state level and managed by individual municipalities and companies. Therefore, any decisions to invest in CNT recovery from batteries will require knowledge of where CNT-enabled batteries will be generated and collected for electronic waste recycling. State level estimates of nanomaterial fate through waste management systems has been reported previously[34] but did not account for potential export or recycling of electronics, with associated risks for occupational exposure and possible recovery and reuse of specific nano-enabled battery materials.

In this section, national results for obsolete portable computers are allocated to state levels. Laptop ownership is clearly a function of affluence but also of demographics and varies widely among age groups. Population levels for different age groups in each state (2012)[35] and nationwide portable computer use percentages for the same age groups provided by Deloitte[36] are used to calculate the number of obsolete portable computers in each state, rather than using GDP and population of each state to allocate the national results to state levels[34]

Statistics on the disposition of electronic waste in each state was used to determine the quantities of CNTs in laptop batteries destined for recycling, incineration, or landfilling. Two scenarios were considered: a baseline scenario assuming current rates of electronic waste recovery for recycling and a scenario in which the United States meets the 85% recovery target put forth in the WEEE directive in the EU by the end of the study period. To date, 25 states have passed laws mandating the recycling of rechargeable batteries and electronic waste, some banning the landfilling of lithium-ion batteries. Quantities of electronic waste collected for recycling in these states has risen steadily.[37,38] State-specific recycling rates for electronic waste were estimated by dividing the quantities collected by national per capita electronic waste generation rates.[39] For states without reported quantities for electronic waste collection, the national average rate of 16.4% was used. The remaining electronic waste was allocated to incineration and landfilling according to the state proportions for regular municipal solid waste in 2008.[40] These waste management rates were then applied to the state-level scenarios for CNTs in laptop batteries. Yearly recycling rates in the second scenario were obtained by setting the 2040 rate at 85% for all states and performing linear interpolation from the current baseline. All waste management rates are given in Tables S1 and S2 of the Supporting Information.

■ RESULTS AND DISCUSSION

Historical Computer Sales Data. Historical data were analyzed to obtain time series estimates for future portable computer sales, penetration rate, and generation of obsolete units.[29] Portable computer sales from 1992 to 2009 are drawn directly from an EPA report.[41] Computer sales data from 2010 to 2012 are calculated based on the sales data for mature markets (Austria, Belgium, The Netherlands, Luxembourg, Denmark, Finland, France, Germany, Italy, Norway, Portugal, Spain, Sweden, United Kingdom, Canada, Japan, and United States) obtained from the International Digital Corporation (IDC).[42] Relative gross domestic product (GDP) and the population of the countries in mature markets are used to estimate the proportion of the mature market sales of portable computers sold in United States.[43] First, the GDP of each country is divided by the highest GDP, and this result is multiplied by the population of each country. Then, the volume of portable computers sold is allocated to each country based on their updated population. According to these estimates, the number of portable computer sales in the United States peaked at approximately 47 million units in 2009, followed by a decrease to 2012 (Figure 1a). The decreasing trend in computer sales between 2010 and 2012 is also observed for other mature markets.

Forecasting of Penetration Rate Using Logistic Model. With the baseline lifespan distribution, historical sales data, and United States population estimates,[44] eqs 1−5 are used to calculate the penetration rate and therefore sales for years 1992 through 2012, by fitting to the historical penetration rates to calculate the constants r (growth rate) and K (demand saturation point).[29,45] The bounding approach suggested by Yang et al.[29] is also used to consider different laptop sales scenarios that represent the lowest and highest conceivable values and to calculate a range of outcomes based on these values.[46]

The upper and lower bounds of the demand saturation point K assumed for personal computers (desktop and portable) by Yang et al.[29] were 1.3 and 1.0, respectively. In contrast, Lam et al.[1] used a logistic model to fit the market share fraction at which portable computers sales saturate the computer market share by 2018; this value was 70%. Also, they considered that portable computers will not penetrate past the modeled saturation level before the year 2030, and there exists an equilibrium saturation of approximately 30% desktops in the computer market for this time frame. On the basis of the upper bounds for PCs and market share percentage, the upper bound value for K is set at 0.8. The optimal value for demand saturation ($K = 0.64$) is calculated using eq 4. The lower bound for K is then assumed to be 0.5, so that the optimal value lies approximately midway between the upper and lower bounds.

Using the estimated values of K, penetration rates N_i were then forecast over the time frame of 2013−2040 using the logistic model described by eq 4. Statistical fits (Anderson−Darling) were performed to determine parameters using upper bounds, optimal value, and lower bound for K mentioned above. The growth rate r and initial penetration rate N_0 are estimated by minimizing the error (λ) for each value of K

$$\lambda = (\widehat{N_t} - N_t)^2 \tag{11}$$

where $\hat{N}_t$ is the penetration rate estimated using the logistic model, and N_t is the actual penetration rate (1992−2012) calculated based on historical data of portable computer sales, population, and lifespan given above. The results of statistical fitting are shown in Table 1.

Table 1. Results of Statistical Fitting to Logistic Model

	upper bound demand saturation ($K = 0.8$)	optimized demand saturation ($K = 0.64$)	lower bound demand saturation ($K = 0.5$)
r	0.196	0.227	0.287
N_0	0.027	0.016	0.007
λ	0.0073	0.0067	0.0083

Optimizing both K and r values offers the best fit of historical data, but all three demand saturation scenarios are considered in the analysis.

Estimation of Prospective Sales and Generation of Obsolete Portable Computers. In 2020, sales of portable computers are estimated to be 66 million for the optimized demand saturation case, with 76 million and 54 million in upper and lower bound scenarios, respectively (Figure 1a). By 2040, 99 million units are estimated to be sold in the upper bound scenario and 63 million in the lower bound scenario, with an optimized scenario of 80 million units. Product obsolescence lags behind initial sales. The annual generation of obsolete portable computers with the consideration of exported used portable computers is shown in Figure 1b. Annual generation of obsolete portable computers will grow from 59 million units in 2020 to 71 million units in 2030 and 76 million units in 2040 (in the optimized middle scenario). The difference between upper and lower bound estimates also increases with time, ranging from 65 to 94 million obsolete units for upper bound and 50−59 million obsolete units for lower bound generated by the end of the study period. The difference between the estimated number of obsolete portable computers generated as electronic waste and the estimated volume of portable computers sold decreases as the number of years considered in the estimation increases.

Technology Penetration of CNT Li-Ion Batteries. The use of a modified logistic model to project baseline scenarios for technology change and replacements for competing electronic products has shown a good fit for desktop-to-laptop computers and CRT-to-LCD display transitions.[1] However, in the case of transition from conventional Li-ion to nano-enabled Li-ion batteries, it is not possible to make a statistical fit given the lack of historical data, as CNT Li-ion batteries have not been extensively commercialized. With the presumption that CNT Li-ion batteries in portable computers will infuse the marketplace, three scenarios are modeled for market fraction growth rate and inflection point parameters to predict penetration: (1) For the low technology substitution case, growth rate (r_n) is 0.4 and inflection point ($t_{n,0}$) is the year 2029. (2) For the medium technology substitution case, growth rate (r_n) is 0.6 and inflection point ($t_{n,0}$) is the year 2024. (3) For the high technology substitution case, growth rate (r_n) is 0.8 and inflection point ($t_{n,0}$) is the year 2020.

The inflection point for each case is the year at which the market fraction of the new technology reaches 50%. As depicted in Figure 2a, a higher growth rate results in a more pronounced S-shape curve. Consistent with an increasing growth rate, inflection points are reached sooner. If the low growth scenario is considered, CNT Li-ion batteries show a 100% market share in approximately 25 years, if the CNT-batteries begin to be commercialized in the next few years. For medium and high growth scenarios, 100% market share would be reached in 18 and 12 years, respectively.

Using prospective sales data obtained with the upper bound value of demand saturation, market share for CNT Li-ion batteries on each year, and baseline lifespan, the volume of obsolete portable computers was estimated for each of the three scenarios for technology substitution. Results for each technology transition scenario and comparison between the total obsolete portable computers and those with nano-enabled batteries are shown for the optimized demand saturation case in Figure 2b. Results for upper and lower bound cases are shown in Figure S1 of the Supporting Information.

In the high technology transition scenario (optimized K value), by 2029, nearly the total number of obsolete portable computers, around 69 million units, will be powered by CNT Li-ion batteries. In the case of medium and low technology transition scenarios, the years at which total obsolete portable computers and obsolete portable computers with nano-enabled batteries curves converge are 2036 and 2040, respectively. As the market share of this type of battery increases, release of CNTs when both portable computers and batteries are disposed could become a major concern. There is uncertainty regarding the behavior that this nanomaterial may have if the

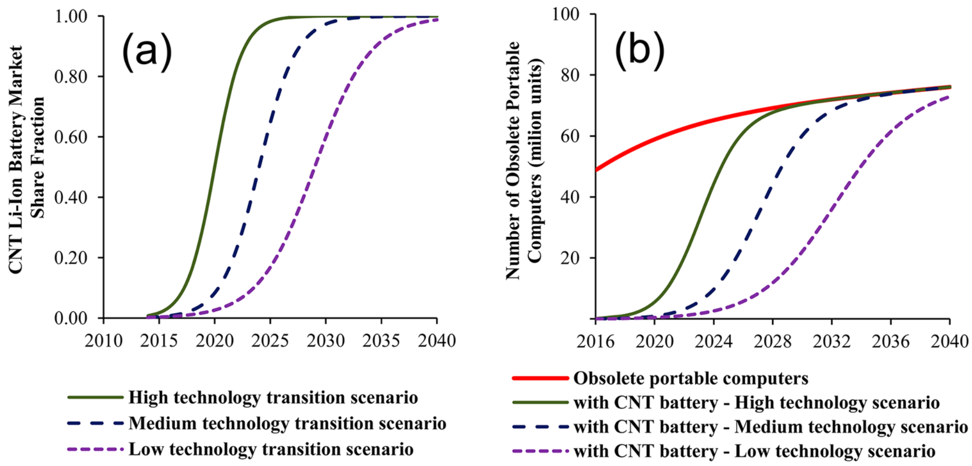

Figure 2. Forecasting (a) market share for conventional Li-ion and CNT Li-ion batteries and (b) estimated generation of total and CNT-containing obsolete portable computers with CNT Li-Ion batteries from 2016 to 2040 (optimized demand saturation).

device that contains it is sent to landfill, incineration, or recycling. As the total number of disposed portable computers increases, recycling infrastructures in the United States might need to increase their capacity and establish new processes to reduce the potential emissions of CNTs.

CNT Mass Flows and End-of-Life Management. Figure 3 shows the amount of MWCNTs that obsolete portable computers with CNTs Li-ion batteries would contain in the United States, as well as the portion of these that are projected to enter landfills.

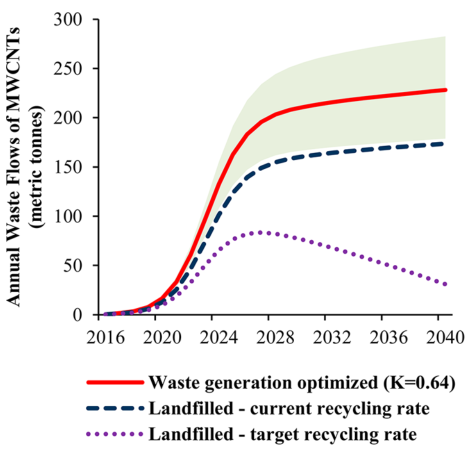

Figure 3. Flows of MWCNTs from obsolete Li-ion batteries entering waste management and landfilled in two scenarios.

The final results for the high technology transition scenario (optimized *K* value) suggest that over the next 25 years, a cumulative 3731 tons of CNTs are expected to be discarded in Li-ion laptop batteries, from a current annual amount of just a few hundred kilograms, and this represents only one application of CNTs. If current recycling practices remain in place, of this cumulative total, 602 tons are projected to be collected, 288 tons incinerated, and 2842 tons landfilled nationally (Table S1, Supporting Information). For context, current use of CNTs in all electronics and optics applications is approximately 800 tons per year.[34] California is expected to collect for recycling the largest amount of CNTs in Li-ion laptop batteries (116 tons) but also landfill the largest amount (330 tons), followed by Texas (290 tons). Incidental incineration of CNT-laden batteries is projected to be greatest in New York (55 tons), Massachusetts (39 tons), and Connecticut (31 tons) over the next 25 years. If the United States achieves an 85% recycling rate, this will reduce the total amount landfilled over the study period by nearly 60% (Figure 3 and Table S2, Supporting Information).

■ UNCERTAINTIES, LIMITATIONS, AND OPPORTUNITIES FOR FUTURE WORK

Technology forecasting is highly uncertain, and the present study makes a number of simplifying, mostly static, assumptions regarding the pace of technology development, consumer behavior, and the management of electronic waste in the United States. In the present study, a discrete probability distribution of lifetimes with average of 3 years for portable computers has been used to estimate both prospective sales and generation of obsolete units. Storage or "hibernation" of obsolete computers certainly takes place, but it is difficult to project into the future and so is not included in the lifetime estimates where it would delay the time to waste management. Reuse is also not considered, as the batteries are often replaced during refurbishment. In addition, in this study, it is assumed that there is no battery replacement during the lifespan of portable computers. The amount of CNTs generated due to the discarded CNT lithium ion batteries would be larger if a battery replacement option was considered in the analysis. Due to accelerated development in computer technology, the average lifespan might decrease (or increase if computers are designed for upgradeability in the years to come). The quantities of CNTs assumed per Li-ion battery cells are fixed at 1 g based on current device fabrication techniques and energy density requirements. However, material efficiency may increase in the future, while battery requirements will almost certainly become more demanding, so it is unclear how the mass of CNTs in each computer will change in the future. A decrease in the amount of CNTs in lithium ion batteries due the technological development in the battery industry, the mass of CNTs generated over time would have a decreasing growth trend or even a decreasing trend over time.

In spite of the robustness that the logistic model has shown for many technology transitions throughout history, market disruptions or the way that computer technology evolves might make the use of a logistic model inappropriate. This is observed

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already in the volatility of recent per capita sales estimates, most likely due to the economic downturn at the end of the 2000s but clearly lacking the monotonic form of a logistic model. Disruption by competing technologies also carries uncertainty. In the past few years, commercialization of tablets has started to grow; this type of device offers many applications and functions that are found in portable computers. Their adoption and substitution for laptops in the consumer electronics market will certainly influence the results presented here, although Li-ion batteries may also be well-suited to tablet computers. Wearable or even implanted computers may become commonplace, and of course battery technology will continue to advance and CNT-based electrodes may be supplanted by superior technology in the future.

On the basis of the current state of technology, however, the scenario results presented here provide useful projections of the quantities and likely location of CNTs generated for an important component of electronic waste, which is information that can be used to plan investments in collection and recycling efforts and ensure safe and responsible handling and disposal measures.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Results for the future generation of obsolete computers under alternate demand saturation assumptions, as well as for the state-level generation and management of CNTs in end-of-life laptop batteries. 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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