Spatio-temporal variation of landfill gas in pilot-scale semi-aerobic and anaerobic landfills over 5 years

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ARTICLE INFO
Article history:
Received 29 June 2016
Revised 19 September 2016
Accepted 23 September 2016
Available online 6 November 2016

Keywords:
Landfill gas
Semi-aerobic landfill
Anaerobic landfill
Municipal solid waste

ABSTRACT
Variation of CH₄, CO₂, and O₂ concentrations in layers of different depths in semi-aerobic and anaerobic landfills was analyzed over a period of 5 years. The results showed that most of the municipal solid waste became basically stable after 5 years of landfill disposal. In the upper and middle layer, the concentration of CH₄ in the semi-aerobic landfill was significantly lower than that in the anaerobic landfill in different landfill periods, while in the lower layer, there was little difference in the CH₄ concentration between the semi-aerobic and anaerobic landfills. The average concentration of CH₄ and CO₂ in the anaerobic landfill was always higher than that in the semi-aerobic landfill, while the O₂ concentration showed an opposite variation in different landfill periods. This was related to the aerobic reaction of landfill waste around the perforated pipe in the semi-aerobic landfill, which inhibited effective landfill gas generation.

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Introduction
The conventional disposal methods for municipal solid waste (MSW) are generally sanitary landfilling, composting and incineration. Because of its advantages of large disposal capacity, convenient operation, easy maintenance, and low investment cost, sanitary landfilling has been the primary method of MSW disposal. Although incineration has recently been receiving increased attention, landfilling is the primary method of MSW disposal because of the current situation in China. Sanitary landfilling can be divided into anaerobic landfilling, aerobic landfilling and semi-aerobic landfilling based on the concentration of oxygen (O₂) in landfill waste layers. Anaerobic landfilling is widely applied because of its advantages including low cost, simple operation, convenient management, and less strict requirements for waste components. However, anaerobic landfilling requires a great quantity of soil and a long stabilization time. In addition, inadequate processing of the leachate generated can easily cause secondary pollution (Read et al., 2001). Compared with anaerobic landfilling, semi-aerobic landfilling can greatly accelerate the stabilization of MSW, reduce the concentration of organic pollutants in the leachate, and reduce landfill gas generation (Aziz et al., 2010). Thus, semi-aerobic landfilling has been widely applied in many developing countries and regions in the last few decades. In recent years, semi-aerobic landfill technology has been promoted widely in China, with the promulgation and operation of a new standard for pollution control on municipal solid waste landfill sites (GB16889-2008, replacing GB16889-1997). At present, there are dozens of semi-aerobic landfills in small- and medium-sized cities such as Weifang, Mengzhi, and Langfang in China.

Since semi-aerobic landfill technology was first proposed, many domestic and overseas scholars have conducted a great
deal of research on the topic, including structure and function (Tanaka, 1993), leachate treatment (Renou et al., 2008; Othman et al., 2010), landfill waste stabilization (Shimaoka et al., 2000), leachate recirculation (Wang et al., 2006), airflow rate, and aerobic radius (Tanaka et al., 1997). At the same time, researchers have begun to pay attention to landfill gas generation in semi-aerobic landfills. The main components of landfill gas are CH$_4$ and CO$_2$. CH$_4$ liberated from landfills is a serious threat to the environment, as its global warming potential is more than 20 times that of CO$_2$ (Kumar et al., 2004). Aronica et al. found that 1 ton of MSW can produce 39–390 m$^3$ landfill gas throughout the life of an anaerobic landfill (Aronica et al., 2009). Thus, landfills are an important emission source of CH$_4$. Matsufuji et al. reported that, according to a model estimation, the contribution of landfill gas to the greenhouse effect can be reduced by 45% in semi-aerobic landfills compared with anaerobic landfills (Matsufuji et al., 1993). Another study showed that CO$_2$ generation in semi-aerobic landfills was higher than that in anaerobic landfills, while CH$_4$ generation was significantly reduced, and the gasification rate (ratio of total landfill gas generation and organics in MSW) in semi-aerobic and anaerobic landfills was 37% and 15%, respectively (Matsufuji and Tachifuji, 1997). Huang et al. found that the CH$_4$ production rate under leachate recirculation was two times that without leachate recirculation in a semi-aerobic landfill (Huang et al., 2008). Dong et al. (2005) and Zhang et al. (2006) both found that the spatial distribution of O$_2$ and CH$_4$ was correlated with the airways in semi-aerobic landfills, and the O$_2$ concentration in semi-aerobic landfills was significantly higher than that in anaerobic landfills, while the CH$_4$ concentration showed the opposite trend.

The application of semi-aerobic landfill technology is not yet mature, and many problems and shortcomings are encountered.

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Fig. 1 – Schematic of the semi-aerobic landfill (a) and anaerobic landfill (b). (1) HDPE membrane; (2) air; (3) leachate collection pipe; (4) landfill storage; (5) plastic membrane; (6) perforated pipe; (7) stone cage; (8) gravel; (9) landfill layer; (10) sampling points of mineralized waste; (11) valve. And the horizontal distribution of landfill gas sampling points in semi-aerobic landfill (c) and anaerobic landfill (d).
in practical application, especially the control of landfill gas emission. There have been few comparative studies of landfill gas emission in semi-aerobic and aerobic landfills over the long term. Therefore, in this study, experiments comparing semi-aerobic and anaerobic landfilling of MSW were conducted at the pilot-scale. In addition, the release characteristics of landfill gas over a 5-year period were analyzed, which provided a theoretical basis for landfill gas emission reduction.

1. Materials and methods

1.1. Pilot-scale landfill device

The size of the semi-aerobic landfill and anaerobic landfill devices was 21 × 3.8 × 6.0 m (Fig. 1a–b). In the bottom of the semi-aerobic and anaerobic landfill bodies, a leachate collection pipe (20 cm × 22 m) was placed above the high-density polyethylene (HDPE) membrane. A stone cage (thickness of 0.6 m) was placed over the leachate collection pipe for protection. In the semi-aerobic landfill, three perforated pipes (15 cm × 6 m) were vertically installed at 1/4, 2/4, and 3/4 along the length of the leachate collection pipe, covered with a stone cage (thickness of 0.4 m) for protection and prevention of air intrusion. Branch pipes for leachate collection were laid in the anaerobic landfill instead of perforated pipes, and a ball valve was installed at the outlet of the main leachate collection pipe to prevent leachate outflow.

The pilot-scale semi-aerobic landfill and anaerobic landfill experiments were conducted simultaneously in Jiujiang, China over a period of 5 years. After MSW was dumped and compacted to 1.5, 3.0, and 4.5 m, sampling devices for landfill gas were set up in the upper, middle, and lower layers of the landfill bodies, respectively (Fig. 1c–d). In the semi-aerobic landfill, 27 sampling points for landfill gas were located in the physical center of the landfill at some distance from the perforated pipe. In the anaerobic landfill, 9 sampling points were spread over the physical center. When MSW was filled and compacted to a height of 5.5 m, it was covered with clay (thickness of 0.3 m) and plastic membrane to ensure sealing of the landfill body. The compaction density of landfill MSW was controlled at about 0.5 × 10^3 kg/m^3.

1.2. Experimental method

Landfill waste was collected from municipal solid waste of Jiujiang, China, and the physicochemical properties are as follows: kitchen garbage 50.84%, vegetation 2.30%, paper 4.56%, fabric 1.20%, plastic 8.74%, brick 3.83%, glass 1.55%, metal 0.18%, lime soil 26.77%, and bulk density 0.52 T/m^3.

According to previous studies (Liu et al., 2005), maximal production of landfill gas appeared 6–12 months after landfilling began in the semi-aerobic landfill. Therefore, in this study, collection and analysis of landfill gas was conducted in the period of initial landfilling (November 2010), period of peak gas generation (November 2011), and period of stable landfilling (November 2015). A portable infrared 4-component gas analyzer with a measurement accuracy of 0.5% (volume proportion) was used to analyze the concentrations of O₂, CH₄, and CO₂ in both semi-aerobic and anaerobic landfills. The sampling frequency was once per week, and the average of values measured four times in a month was used as the concentration of landfill gas for that month.

The manual sorting and weighing method was adopted to determine the components of mineralized waste in the semi-aerobic and anaerobic landfills. The mineralized wastes from different sampling points were uniformly mixed after stratified sampling. The specific distribution of sampling points is shown in Fig. 1. The collected samples (500 g) were taken back to the laboratory for analysis. According to standard CJ/T99-1999, pH, total nitrogen (TN), and total phosphorus (TP) of mineralized waste was determined using glass electrode method, semi-micro Macro Kjeldahl method and ammonium molybdate spectrophotometric method, respectively. Moisture content was measured using method of sampling and physical analysis of domestic waste according to standard CJ/T313-2009.

2. Results and discussion

2.1. Physical and chemical properties of mineralized waste

The composition of mineralized waste in the anaerobic landfill and semi-aerobic landfill is shown in Fig. 2. The main component of MSW was kitchen garbage, followed by lime soil.

Fig. 2 – Composition of mineralized waste in anaerobic landfill and semi-aerobic landfill.
After 5 years of semi-aerobic and anaerobic landfill disposal, kitchen garbage and other easily biodegradable organics in MSW had been completely degraded. Lime soil was the largest component in mineralized waste. There was little difference in the composition of mineralized wastes between the anaerobic landfill and semi-aerobic landfill. Moreover, during mineralized

<table>
<thead>
<tr>
<th>Sample</th>
<th>Moisture content</th>
<th>pH</th>
<th>Conductivity (μS/cm)</th>
<th>Organic matter (g/kg)</th>
<th>TN (g/kg)</th>
<th>TP (g/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mineralized waste of semi-aerobic landfill</td>
<td>32.5%</td>
<td>7.6</td>
<td>1543.0</td>
<td>158.1</td>
<td>8.9</td>
<td>7.2</td>
</tr>
<tr>
<td>Mineralized waste of anaerobic landfill</td>
<td>34.0%</td>
<td>7.4</td>
<td>2060.0</td>
<td>170.6</td>
<td>10.0</td>
<td>9.9</td>
</tr>
</tbody>
</table>

Fig. 3 – Spatio-temporal variation in landfill gas in the upper layer of the semi-aerobic and anaerobic landfills (landfill depth: 4.5 m). (I)—the initial landfill period; (II)—the peak gas-generating period; (III)—the stable landfill period.
waste sample collection, most of the mineralized waste appeared as dark brown quasi lime soil with loose structure, and without peculiar smell. It differed greatly from the appearance of fresh waste. This indicated that fresh waste had been rendered basically stable after 5 years of landfill disposal.

The physicochemical properties of mineralized waste are shown in Table 1. The pH of mineralized waste was weakly alkaline. In addition, the conductivity of mineralized waste in the semi-aerobic landfill was significantly lower than that in the anaerobic landfill. The organic matter, TN, and TP of mineralized waste were in accordance with the nutrient requirements of urban wastes for agricultural control in China (GB8172-87) (organic matter ≥10%; TN ≥ 0.5%; TP ≥ 0.3%), indicating its land-use potential.

2.2. Variation of landfill gas concentration in different landfill layers of semi-aerobic and anaerobic landfills over 5 years

2.2.1. Upper layer (burial depth = 4.5 m)
Spatio-temporal variations of landfill gas and the average gas concentration in the upper layer of the semi-aerobic landfill and anaerobic landfill are shown in Figs. 3 and 4, respectively.

The O2 concentration in the upper layer of the semi-aerobic landfill showed an increasing trend and was maintained at a relatively high level. Specifically, it increased from 9.16% in the initial landfill period to 13.71% in the peak gas-generating period, and then further increased to 19.07% in the stable landfill period, which was close to the concentration of O2 in the air (21.0%). The O2 concentration in the upper layer of the anaerobic landfill, which had been kept under anaerobic conditions, was relatively low in the initial landfill period and peak gas-generating periods (1.25% and 1.03%, respectively). After 5 years, the O2 concentration in the upper layer of the anaerobic landfill had increased significantly to 12.92%, which indicated that the upper landfill layer had become completely aerobic. This may be related to air intrusion due to the settlement process of landfill waste caused by organic matter degradation and damage to the monitoring pipe, which resulted in the presence of local areas with higher O2 concentration (Swati and Joseph, 2008; Zhang et al., 2010).

In general, the O2 concentration in the upper layer of the semi-aerobic landfill showed an increasing trend and was maintained at a relatively high level. Specifically, it increased from 9.16% in the initial landfill period to 13.71% in the peak gas-generating period, and then further increased to 19.07% in the stable landfill period, which was close to the concentration of O2 in the air (21.0%). The O2 concentration in the upper layer of the anaerobic landfill, which had been kept under anaerobic conditions, was relatively low in the initial landfill period and peak gas-generating periods (1.25% and 1.03%, respectively). After 5 years, the O2 concentration in the upper layer of the anaerobic landfill had increased significantly to 12.92%, which indicated that the upper landfill layer had become completely aerobic. This may be related to air intrusion due to the settlement process of landfill waste caused by organic matter degradation and damage to the monitoring pipe, which resulted in the presence of local areas with higher O2 concentration (Swati and Joseph, 2008; Zhang et al., 2010).

There was also a sustained increase of O2 concentration in the semi-aerobic landfill, while an obvious increase was only observed in the stable landfill period in the anaerobic landfill.

The CH4 concentration in the upper layer of the anaerobic landfill reached a clear peak of gas generation after 1 year, while there was no significant peak of CH4 gas generation in the semi-aerobic landfill. The CH4 concentration in the anaerobic landfill was 11.90% and 26.65% in the initial landfill period and peak gas-generating period, respectively, which was significantly higher than that in the semi-aerobic landfill (4.13% and 3.36%, respectively). This was because the O2 content and aerobic area close to the perforated pipe was richer and larger, leading to inhibition of CH4 production (Chun, 2014). After 5 years, almost no CH4 was detected in the upper layer of the semi-aerobic landfill, while it was relatively low in the anaerobic landfill (0.58%), which indicated that the mineralized waste was basically stable. In general, the concentration of CH4 in the upper layer of the semi-aerobic landfill was significantly lower than that in the anaerobic landfill in the early stage of the landfill. Moreover, the CH4 concentration showed a clear spatial distribution in the upper layer of the semi-aerobic landfill, which indicated that the design of the perforated pipe in the semi-aerobic landfill could markedly inhibit CH4 production (Shi et al., 2014).

The CO2 concentration in the upper layers of both the semi-aerobic landfill and anaerobic landfill gradually decreased with landfilling time. The CO2 concentration in the upper layer of the semi-aerobic landfill varied widely, falling from 12.28% in the initial landfill period to 6.89% in the peak gas-generating period, and to 0.60% in the stable landfill period. In the anaerobic landfill, CO2 was relatively high and uniform in the initial period and peak gas period (33.14% and 27.02%, respectively). During the stable landfill period, the CO2 concentration decreased to 5.12%. Accordingly, the CO2 concentration in the upper layer of the anaerobic landfill was significantly higher than that in the semi-aerobic landfill in each corresponding landfill period. This was mainly related to the semi-open system of the semi-aerobic landfill, which resulted in faster exchange of generated CO2 with the outside air (Matsuto et al., 2015). Thus, the semi-aerobic landfill more easily attained a steady state. Similar to the distribution of CH4 concentration, the airway design also led to a CO2 concentration with an apparent spatial distribution.

2.2.2. Middle layer (burial depth = 3.0 m)
The spatio-temporal variation of landfill gas and average gas concentration in the middle layer of the semi-aerobic landfill and anaerobic landfill are shown in Figs. 5 and 6, respectively.

In the middle layer of the semi-aerobic landfill, the O2 concentration was relatively low in the initial landfill period (2.73%) and the peak gas period (2.11%). Thus, at this time, the middle layer was in a facultative anaerobic condition. There was an obvious spatial distribution of O2 concentration. During the stable landfill period, the entire middle layer of the semi-aerobic landfill maintained aerobic conditions with relatively high O2 concentration (17.52%). These findings imply that there was less consumption of O2 in the middle layer of the semi-aerobic landfill (Park et al., 1997).
Throughout the anaerobic landfill period, the \( O_2 \) concentration increased after slightly decreasing in the early landfill period. Because of the intrusion of \( O_2 \) during landfill operation, the middle layer of the anaerobic landfill in the initial period maintained facultative aerobic conditions. Then due to the \( O_2 \) consumption caused by MSW decomposition, the \( O_2 \) concentration in the peak gas-generating period was very low (0.52%), corresponding to fully anaerobic conditions. In the stable landfill period, there was an obvious increase in \( O_2 \) concentration (7.15%). At this time, most regions remained anaerobic, although some regions were still partially aerobic. In general, the \( O_2 \) concentration in the middle layer of the semi-aerobic landfill during each landfill period was significantly higher than that in the anaerobic landfill, and an
obvious increase was observed at the end of the period in both landfills.

There was no obvious peak of CH4 gas generation in the middle layer of the semi-aerobic landfill, while the opposite was found in the anaerobic landfill. In the initial landfill period and the peak gas-generating period, the CH4 concentration was relatively high (13.71% and 12.56%), and its distribution was uniform throughout the middle layer. During the stable landfill period, CH4 was only detected at a few gas sampling points. These findings indicated that at this time the landfill waste had been rendered relatively stable. The CH4 concentrations were 12.71%, 38.20%, and 2.61% in the initial landfill period, peak gas period, and stable landfill period, respectively in the anaerobic landfill. In general, the CH4 concentration in the middle layer of the semi-aerobic landfill in the early landfill stage was significantly lower than that in the anaerobic landfill. These results indicated that an obvious spatial distribution and a relatively uniform distribution of CH4 appeared in the semi-aerobic landfill and anaerobic landfill, respectively.

Variations in CO2 concentrations in the middle layer of the semi-aerobic and anaerobic landfills were similar to those in the upper layer, showing a decrease along with landfill time. During the initial landfill period, peak gas-generating period, and stable landfill period, the CO2 concentrations in the middle layer of the semi-aerobic landfill were 23.84%, 19.82%, and 1.50%, respectively, while in the anaerobic landfill they were 41.03%, 32.35%, and 9.53%, respectively. It can be inferred that landfill waste stability was significantly higher in the middle layer of the semi-aerobic landfill than that in the anaerobic landfill.

2.2.3. Lower layer (burial depth = 1.5 m)
The spatio-temporal variations in landfill gas and average gas concentration in the lower layer of the semi-aerobic landfill and anaerobic landfill are shown in Figs. 7 and 8, respectively.

Over the landfill period, the O2 concentration in the lower layer of the semi-aerobic landfill decreased from 3.52% in the initial landfill period to 1.97% in the peak gas period, and then significantly increased to 9.08% in the stable landfill period. In the stable period, the lower layer of the semi-aerobic landfill was aerobic. The O2 concentration in the lower layer of the anaerobic landfill showed a gradually decreasing trend. In the initial landfill period, the lower layer of the anaerobic landfill was under facultative aerobic conditions because of the low O2 concentration (2.96%). In the peak gas period and the stable landfill period, the O2 concentration in the lower layer of the anaerobic landfill was very low (0.39% and 0%), suggesting that the lower layer of the landfill was completely anaerobic. In general, the O2 concentration in the lower layer of the semi-aerobic landfill in each period was significantly higher than that in the anaerobic landfill, leading to great differences in the degree of stabilization of landfill waste at the bottom.

Variations in CH4 generation were very similar between the semi-aerobic landfill and the anaerobic landfill. During the initial landfill period, peak gas period, and stable landfill period, the CH4 concentrations in the lower layer of the semi-aerobic landfill were 15.63%, 34.87%, and 8.49%, respectively, while they were 14.49%, 32.76%, and 10.56%, respectively, in the anaerobic landfill. There was little difference in the CH4 concentration between the semi-aerobic and anaerobic landfills during each corresponding landfill period. This indicated that the perforated pipe design in the semi-aerobic landfill did not have an obvious effect on aerobic degradation of landfill waste in the lower layer of the landfill (Filipkowska, 2008). The inhibitory effect on CH4 production due to the semi-aerobic landfill structure primarily occurred in the upper and middle layers and around the airway, while the landfill gas reduction effect at the bottom of the landfill was not obvious.

Variations in CO2 concentration in the lower layer were similar between the semi-aerobic landfill and anaerobic landfill, showing a decreasing trend along with landfill time. In the lower layer of the semi-aerobic landfill, the CO2 concentration was 31.71%, 30.06%, and 10.27% in the initial landfill period, peak gas period, and stable landfill period, respectively, while in the anaerobic landfill it was 37.51%, 30.93%, and 18.04%, respectively. These findings indicate that biochemical reactions in the lower layer of the semi-aerobic and anaerobic landfills were still occurring.

2.3. Variations in landfill gas during different periods of semi-aerobic and anaerobic landfills

The average variations in concentrations of landfill gas in the semi-aerobic and anaerobic landfills are shown in Fig. 9. The concentration of CH4 in the semi-aerobic and anaerobic landfills increased from 9.58% and 14.81% in the initial landfill period to 17.17% and 30.58% in the peak gas period, respectively. The growth rate reached 79.23% and 106.48%, respectively. After 5 years, the CH4 concentration in the semi-aerobic and anaerobic landfills decreased significantly to 2.82% and 5.26%, respectively. These findings indicate that the CH4 concentration in the anaerobic landfill was significantly higher than that in the semi-aerobic landfill in each corresponding landfill period. This was primarily because of the aerobic and facultative anaerobic areas appearing in the semi-aerobic landfill, which could inhibit the generation of CH4.

During the initial landfill and peak gas period, O2 concentrations remained at a relatively high level in the semi-aerobic landfill (5.22% and 6.81%), while being 2.23% and 0.61% in the anaerobic landfill, respectively. These findings indicated that the semi-aerobic and anaerobic landfills were in aerobic and anaerobic fermentation states, respectively. As the landfill
process continued, the aerobic scope in the semi-aerobic landfill expanded further, and the $O_2$ concentration increased significantly. This may have been owing to the degradation of organic components such as kitchen garbage and paper, which resulted in a relatively loose landfill waste structure, contributing to $O_2$ intrusion. The $O_2$ consumption by mineralized waste was also significantly reduced. After 5 years, the $O_2$ concentration in the semi-aerobic landfill was 15.14%, while it was only 5.09% in the anaerobic landfill. In general, the $O_2$ concentration in the semi-aerobic landfill was
significantly higher than that in the anaerobic landfill at different landfill stages.

The variations in CO₂ concentrations between semi-aerobic and anaerobic landfills were similar. CO₂ declined continuously from the values of 21.57% and 37.26% in the initial landfill period to 4.13% and 11.98% in the stable landfill period. The organic component of landfill waste can generate CO₂ and H₂O through aerobic reaction, while it can generate CH₄, CO₂, and H₂O through anaerobic reaction (Kumar et al., 2016). The decreasing CO₂ concentration reflected the continuous decomposition of organic matter in the landfill process. The concentration of CO₂ in the semi-aerobic landfill was significantly lower than that in the anaerobic landfill in each corresponding landfill period. This was mainly related to the semi-open condition in the semi-aerobic landfill and the fully closed condition in the anaerobic landfill. The semi-open system was beneficial to the intrusion of O₂ and spread of CO₂ and CH₄.

Comparison of the generation of landfill gas in semi-aerobic and anaerobic landfills shows that the semi-aerobic landfill was more conducive to reducing the generation of CH₄ and CO₂ in the long term, which has great advantages for the reduction of greenhouse gas emissions.

3. Conclusions

After 5 years of landfill disposal, most of the mineralized waste in semi-aerobic and anaerobic landfills remained basically stable. There was little difference in the composition and physicochemical properties of mineralized waste between the semi-aerobic landfill and anaerobic landfill.

In the upper and middle layers, the concentration of CH₄ in the semi-aerobic landfill was significantly lower than that in the anaerobic landfill in different landfill periods, while in the lower layer, there was little difference in the CH₄ concentration between the semi-aerobic landfill and anaerobic landfill. The main CH₄ emissions in the semi-aerobic landfill occurred in the lower layer. The CO₂ concentrations in different layers of the semi-aerobic landfill were higher than in the anaerobic landfill, while the concentration of O₂ showed the opposite variation.

The average concentrations of CH₄ and CO₂ in the anaerobic landfill were always higher than those in the semi-aerobic landfill in each corresponding landfill period. This was related to the aerobic reaction of landfill waste around the perforated pipe in the semi-aerobic landfill, which inhibited the CH₄ generation.

Acknowledgments

This work was supported by the National Science and Technology Support Program Project (No. 2014BAL02B01). We thank Prof. Dong Lu and He Jie for assistance with the field experiments. We thank Jeremy Kamen for help in writing the article.

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