



Article Investigation on the Emission and Diffusion of Hydrogen Sulfide during Landfill Operations: A Case Study in Shenzhen

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Abstract: This study investigated the emission and diffusion of hydrogen sulfide (H₂S), as one of the odorous gases generated from landfills, in a municipal solid waste landfill of a south Chinese city. To this end, the flux of the H₂S emissions in the working area of the landfill and its diffusion in the surrounding area were measured. The diffusion of the H₂S was simulated at different wind speeds, wind directions, bare working areas of the landfill, heights of the landfill, and angles between the wind direction and the long side of the working area. The results indicated that the concentration of the H₂S around the monitoring point ranged from 0 to 60 μ g/m³, and the simulated data were consistent with the measured results. At a uniform wind direction, the pollution range of the H₂S was narrow. Furthermore, with an increase in the height of the waste dump, the concentration of the H₂S decreased in the working area and the wind direction was 0°, the H₂S largely spread along the extension cord of the long side of the working area. When the angle increased to 90°, the influence range of the H₂S extended significantly. The working area in the landfill site should be regulated based on the monitored data to reduce the effect of this harmful gas on the living environment, and the health of the landfill staff and nearby residents.

Keywords: landfill; hydrogen sulfide; diffusion; in-situ monitoring; static chamber; CALPUFF model

1. Introduction

The emission of odorous gases has become a major concern in both developed and developing countries around the world. In China, particular attention has been paid to the disposal of municipal solid waste (MSW) due to the rapid development of urbanization [1–5]. For its large capacity, low treatment cost, high rate of land utilization, and convenient management, large-scale sanitary landfill is widely employed in China to treat MSW [6]. However, landfill gases (LFGs), which are harmful to the environment, are generated during biochemical reactions in landfills. LFGs are mainly composed of 45–60% methane, 40–60% carbon dioxide, and a small number of odorous gases [7,8]. The formation and diffusion of odorous gases results in significant air pollution in the landfill and its surrounding area, seriously affecting the quality of life and the health of nearby residents [9–11].

LFGs are produced during the biochemical degradation of MSW. Although the main components of LFGs are methane and carbon dioxide, other trace toxic and harmful gases



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are also included, where hydrogen sulfide is one of the essential odorous gases [7,12,13]. Human beings are susceptible to hydrogen sulfide because it has a low olfactory threshold of 0.62 μ g/m³, with a typical rotten egg smell. High concentrations of hydrogen sulfide can affect the human nervous and respiratory systems. For instance, it can paralyze the sense of smell at a concentration of more than 7.59 \times 10⁴ μ g/m³. Moreover, hydrogen sulfide can cause instant death at a concentration of higher than 1.52 \times 10⁶ μ g/m³. The Chinese National Standard (GBZ2.1-2019) stipulates that the maximum allowable concentration of hydrogen sulfide should be lower than 1.52 \times 10⁴ μ g/m³.

So far, research into landfill odor has mainly focused on the distribution and diffusion of volatile organic compounds in the landfill and the surrounding area [7,9,10,14], and there are few studies on the diffusion of hydrogen sulfide in the landfill and the surrounding area. Panza and Belgiorno, 2010 [15], found that construction waste with gypsum walls could produce high concentrations of hydrogen sulfide, in the range of $1.06 \times 10^4 \,\mu\text{g/m}^3$ to $1.52 \times 10^6 \,\mu\text{g/m}^3$. Heaney et al., 2011 [16], studied the relationship between the odor, the concentration of hydrogen sulfide, and the health of the residents living near the landfill. The results show that the stench of the landfill was related to the concentration of hydrogen sulfide when the wind blew from the landfill to the community. Considering the human body's high sensitivity to hydrogen sulfide, it is worth further field testing and studying the distribution and diffusion of hydrogen sulfide in and around landfills, which is of great significance to the life quality and health of the landfill staff and the residents living in the surrounding area.

An atmospheric diffusion model, based on data of meteorological, terrestrial, and odor emissions, is a practical method to simulate how the odorous gas diffuses into the atmosphere [17,18]. The CALPUFF modeling system is one of the most commonly employed atmospheric diffusion models, which assumes that the concentration distribution in a plume cross-section follows a Gaussian curve. The model has been utilized to simulate the chemical transport and dispersion of pollution from chimneys, landfills, vehicles, and ponds, within a range of tens of kilometers [7,19,20]. According to the results of related research concerning the odorous gas diffusion [9,21–23], the effects of odorous gas on the surrounding area are closely related to odor emissions, meteorological and terrestrial conditions, and the distance from the source of pollution. However, the corresponding investigations have mainly focused on the diffusion of volatile organic compounds [24–27]. Therefore, there is still a lack of comprehensive studies on how hydrogen sulfide distributes and diffuses into the landfill and its surrounding area.

As the MSW in the landfill operation area is exposed to the air during operation, a large amount of odor is discharged from the area. Under such circumstances, it is necessary to determine the emission intensity and the diffusion of hydrogen sulfide during landfill operations. Thus, this study measured and analyzed the emissions and diffusion of hydrogen sulfide in a landfill in south China during landfill operations. The key factors affecting the diffusion of hydrogen sulfide were also discussed. To this end, the static chamber method was employed to measure the flux of the hydrogen sulfide emissions in the working area of the landfill, and the measured flux was then used as the initial intensity of hydrogen sulfide pollution emission source, and input into the CALPUFF model to simulate the diffusion of the hydrogen sulfide. Then, the simulated data were compared with the results measured around the landfill. Furthermore, the impacts of different wind speeds, wind directions, bare working areas of the landfill, heights of the landfill, and angles between the wind direction and the long side of the working area were simulated. Thus, the critical factors affecting the diffusion of the hydrogen sulfide were determined and the underlying mechanism of the diffusion of hydrogen sulfide in the landfill was illuminated.

2. Materials and Methods

2.1. Landfill Description

The landfill as the first modern large-scale sanitary landfill in China is an important infrastructure for the disposal of the local MSW (see Figure 1). It covers a total area of 145 hectares and was launched in October 1997. This landfill is designed with a total storage capacity of 46.93 million m³, and a total service life of more than 30 years. It has an average daily waste disposal capacity of 6800 tons, accounting for about 26% of the city's total waste. The construction of the landfill includes three phases. The first phase of the project was built in Xiaping valley with an area of 63.4 hectares, a designed capacity of 14.93 million m³, and a service life of 12 years. The second phase of the landfill will be constructed in the upper and lower plateau valley in the upstream section of Xiaping, with an area of 55.8 hectares, a designed capacity of 12 million m³, and a service life of 10 years. The third phase will heighten the upper part of the first and the second phases by 50–60 m, with a design capacity of 20 million m³.



Figure 1. A satellite image of the landfill.

Based on the climatic conditions of the landfill area in 2018, the meteorological data of January, April, July, and October represented the typical meteorological conditions of winter, spring, summer, and autumn, respectively. Thus, the assessment of the hydrogen sulfide diffusion was simplified to four months: January, April, July, and October. Figure 2 represents a wind rose chart composed of the wind direction and wind speed recorded by local weather stations in 2018. As shown in Figure 2, the wind speed and the wind direction in January, July, and October were relatively concentrated, and the wind chiefly blew from the east at a wind speed in the range of 4 to 7 m/s. Notably, the wind speed and the wind direction were unstable in April. In fact, the wind blew from the northeast (approximately 24%) and the southwest (approximately 22%) at a wind speed largely in the range of 4 to 11 m/s. Only the southwesterly wind had a speed greater than 11 m/s. Generally, the wind speed in the studied area ranged from 0.5 to 11 m/s, and the easterly wind was dominant throughout 2018. Therefore, the online monitoring device (Model 450i hydrogen sulfide—sulfur dioxide analyzer, Thermo Fisher Scientific Inc., USA) for measuring the concentration of the hydrogen sulfide was installed in the northwest area of the landfill (see Figure 1), which was in the downwind direction. The concentration of hydrogen sulfide was measured at hourly intervals, and the data were recorded and uploaded to the background.



Figure 2. Wind roses of the landfill site in the four seasons: (a) January; (b) April; (c) July; (d) October.

2.2. Gas Flux

In this work, the static chamber method was used to measure the flux of the hydrogen sulfide emissions [7]. This method was proposed by Hutchinson and Mosier, 1981 [28], and has been widely employed for the measurement of gas emissions from landfills. The static chamber device used herein was made of plexiglass with a height of 550 mm and a diameter of 400 mm, as depicted in Figure 3. In order to inhibit gas exchange inside and outside of the chamber, it was placed on a metal base buried in the working area to a depth of 10–20 cm, and sealed with water in the contact area during the measurement of the flux of the hydrogen sulfide emissions. A miniature fan was placed inside the chamber to circulate the gas and maintain its uniformity. Through the gas sampling port on the top of the static chamber and the air pump, a gas sample was extracted into Tedlar[®] air sample bags once every 0.5 h, four consecutive times. The gas sample was then tested for the concentration of hydrogen sulfide using gas chromatography–mass spectrometry (GC–MS). The flux of the hydrogen sulfide emissions was determined according to Equations (1) and (2):

$$VC_{t+\Delta t} = VC_t + j'S\Delta t \tag{1}$$

$$j' = \frac{V}{S} \frac{dC}{dt} \tag{2}$$

where *V* is the volume of the static chamber (m³); *C*_t and *C*_{t+ Δt} represent the concentration of the target gas at time *t* and *t* + Δt respectively ($\mu g/m^3$); *S* denotes the bottom area (m²);



 Δt stands for the time interval of the test (s); j' indicates the gas flux ($\mu g \cdot m^{-2} \cdot s^{-1}$); and $\frac{dC}{dt}$ is the slope of the concentration accumulation curve.

Figure 3. The static chamber buried in the field test.

The flux of the hydrogen sulfide emissions measured at ambient temperature, T (K), was calculated by Equation (3):

$$j = j' \frac{273}{T} \tag{3}$$

2.3. CALPUFF Model

The CALPUFF model is commonly adopted to simulate the diffusion of specific pollution sources in particular areas under definite meteorological conditions, and is one of the prediction models recommended for environmental impact assessment in China. This model is chiefly composed of three parts. The first part is the meteorological module (Calmet module) which involves the meteorological data and topographic parameters obtained from meteorological stations in a specific region. It combines the topographic and meteorological data organically, divides the grid, and establishes an analysis model. The second part is the simulation computing module (Calpuff module). Based on the meteorological conditions and the analysis model established by the Calmet module, this part simulates the diffusion of specific pollutants. The third part is the post-processing module (Calpost module), through which the simulation results can be output in the form of a table or cloud map as needed. The transport mechanism of pollutants in the CALPUFF model is based on the Lagrangian Gaussian plume model [7,17–19].

In order to study the influence of wind speed and wind direction on the diffusion of hydrogen sulfide, the meteorological conditions in different seasons (i.e., January, April, July and October) were selected as the simulation parameters. Meanwhile, the landfill operation area was 4000 m², the height of working face was 30 m, and the angle between the wind direction and the long side of the working face was 0°. The diffusion of hydrogen sulfide was also simulated under different operation areas (i.e., 4000, 8000, 12,000 and 20,000 m²) to analyze the effect of landfill operation area. The meteorological conditions in January were selected, the height of the working face was 30 m, and the angle between the wind direction and the long side of the working face was 0°. To analyze the impact of landfill height on the diffusion of hydrogen sulfide, landfill heights of 25, 50, 75, and 100 m were selected as simulation parameters. The meteorological conditions in January were selected area was 4000 m², and the angle between the wind direction and the long side of working face was 0°. Moreover, for studying the influence of the angle between wind direction and the long side of working face, the diffusion of hydrogen sole of working face, the diffusion of hydrogen was 0°.

sulfide at an angle of 0, 45, and 90° between the wind direction and the long side of the working face were simulated respectively. The meteorological conditions in January were selected, the landfill operation area was 4000 m², and the height of the working face was 30 m.

3. Results and Discussion

3.1. H₂S Emissions

Figure 4 shows the variation in the concentration of the hydrogen sulfide near the field boundary based on the data measured by the online monitoring points arranged around the landfill in 2018. The concentration of the hydrogen sulfide around the landfill largely ranged from 0 to 60 μ g/m³, which exceeded the olfactory threshold, thereby causing an intense obnoxious odor. The concentration of the hydrogen sulfide in the landfill in the first half of the year (around 30 μ g/m³) was higher than that in the second half of the year (around 10 μ g/m³), which might be associated with the change of the wind speed and the wind direction in the different seasons. The measured results are consistent with other studies [16,29]. Furthermore, the flux of the hydrogen sulfide emissions measured by the static chamber was input to the CALPUFF model as the intensity of hydrogen sulfide pollution emission source. The simulation results of the diffusion of the hydrogen sulfide are presented in Figure 5. The calculated square areas in the figure, such as the red square, all have a side length of 5 km. The different colors represent various concentrations of the hydrogen sulfide, and the data in Figure 5 denote the maximum concentration of the hydrogen sulfide per hour of the year. The results indicate that the concentration of the hydrogen sulfide near the odor monitoring point was 10–40 μ g/m³, and the simulation data were consistent with the measured results.



Figure 4. The variation in the concentration of hydrogen sulfide with time at the monitoring point.



Figure 5. The diffusion diagram of the hydrogen sulfide based on the measured diffusion flux.

3.2. Influence of Wind Speed and Wind Direction

In order to investigate the influence of the wind speed and wind direction in different seasons on the diffusion range and concentration distribution of the hydrogen sulfide, meteorological conditions in January, April, July, and October were selected as the parameters for analysis. As displayed in Figure 6, the diffusion range of the hydrogen sulfide in the landfill was affected by the wind direction. Due to the influence of the wind, the diffusion range of the hydrogen sulfide was mainly in the downwind direction, while the upwind direction was less affected. It was more likely to cause a bad smell for residents when the wind blew from the landfill to the community [16]. When the wind direction was relatively uniform, the influence range of the hydrogen sulfide was narrow. However, it widened significantly as the wind direction changed greatly. According to the local meteorological data, the wind direction changed significantly and covered nearly 180° in April, and the diffusion area of the hydrogen sulfide was nearly half of the calculated area (Figure 6b). This was consistent with the report of Wang et al., 2019 [7]. Meanwhile, the local meteorological data confirmed that the wind mainly blew from the northeast in October, and the wind direction did not change much. Therefore, the hydrogen sulfide spread over a limited area, accounting for only a quarter of the calculated area (Figure 6d).



Figure 6. Plume trajectories of the hydrogen sulfide under different wind speeds and wind directions in different seasons: (**a**) January; (**b**) April; (**c**) July; (**d**) October.

3.3. Influence of Bare Working Area of Landfills

As shown in Figure 7, the diffusion of the hydrogen sulfide was simulated by considering working areas of 4000, 8000, 12,000, and 20,000 m² to study the influence of different working areas on the diffusion range and concentration distribution of the hydrogen sulfide. The results indicate that the diffusion area of the hydrogen sulfide did not increase markedly, but the concentration of the hydrogen sulfide increased significantly with an expansion of the working area. Simultaneously, the influence range of the hydrogen sulfide was almost similar under different working areas, but its concentration value was different.

When the working area was 8000, 12,000, or 20,000 m², the maximum concentrations of the hydrogen sulfide in the center of the working area were equal to 241, 315, and 447 μ g/m³ respectively, indicating that the concentration of the hydrogen sulfide in the center of the working area correlated positively with the working area [7,29].



Figure 7. Plume trajectories of the hydrogen sulfide under different working areas: (**a**) 4000 m² working area; (**b**) 8000 m² working area; (**c**) 12,000 m² working area; (**d**) 20,000 m² working area.

3.4. Influence of Landfill Height

With an increase in the volume of the landfill, the height of the landfill rises continuously. Figure 8 illustrates the simulated diffusion range and concentration distribution of the hydrogen sulfide under different heights of the landfill. Although the height of the MSW varied, the maximum concentration of the hydrogen sulfide in the landfill occurred in the landfill working area [7]. With an increase in the height of the MSW dump, the concentration of the hydrogen sulfide on the landfill surface gradually declined at the same landfill working area, wind speed, and wind direction, while the concentration of the hydrogen sulfide within the surrounding area increased. At an MSW dump height of 25, 50, and 100 m, the maximum concentration of the hydrogen sulfide in the landfill area was 144, 82, and 27 μ g/m³ respectively. In addition, the concentration of the hydrogen sulfide in the working area decreased with an increase in the height of the MSW dump, but the area with high concentrations of the hydrogen sulfide expanded remarkably. As shown in Figure 8a, the area with a concentration of the hydrogen sulfide in the range of 10–30 μ g/m³ extended remarkably as the height of the MSW dump increased from 25 to 100 m. Indeed, the fluidity of the upper air rose as the height of the MSW dump increased, which caused the hydrogen sulfide in the central area to migrate to the surrounding area. Thus, the diffusion of the hydrogen sulfide was more noticeable.



Figure 8. Plume trajectories of the hydrogen sulfide under different heights of the MSW dump: (a) 25 m; (b) 50 m; (c) 75 m; (d) 100 m.

3.5. Influence of Angle between Wind Direction and Working Area

Figure 9 reveals the diffusion of the hydrogen sulfide at angles of 0, 45, and 90° between the wind direction and the long side of the working area at an identical wind speed, wind direction, bare area of the landfill, and height of the MSW dump. Figure 9a implies the input value of the wind direction and wind speed in the landfill area. The red area in the left corner of Figure 9b-d represents the landfill working area with the dimensions 100 m \times 40 m, and the arrow indicates the wind direction. It can be seen in Figure 9 that the angle between the long side of the working area and the wind direction affected the diffusion of the hydrogen sulfide. In fact, at an angle of 0° (see Figure 9b), the hydrogen sulfide largely diffused along the extension cord of the long side of the working area and thus accumulated in the downwind area significantly [16]. Hence, the influence of the hydrogen sulfide on both sides of the upwind area was negligible. Nevertheless, at an angle of 90° (Figure 9d), the influence of the hydrogen sulfide increased considerably, especially on both sides of the working area. However, at this time, the maximum concentration of the hydrogen sulfide was $40.2 \,\mu g/m^3$, which was lower than the concentration of the hydrogen sulfide at an angle of 0° (65.1 µg/m³). Notably, the influence range of the hydrogen sulfide was larger at an angle of 45° (Figure 9c) than at an angle of 0 and 90°. The highest concentration of the hydrogen sulfide in the center of the working area was also measured at an angle of 45° . Indeed, when the angle between the long side of the working area and the wind direction was 90°, the hydrogen sulfide in the center of the landfill working area was more likely to be affected by the wind. Thus, the hydrogen sulfide was not able to accumulate in the center of the working area.



Figure 9. Plume trajectories of the hydrogen sulfide at different angles between the wind direction and the long side of the working area: (**a**) wind rose; (**b**) an angle of 0 degrees; (**c**) an angle of 45 degrees; (**d**) an angle of 90 degrees.

4. Conclusions

This study employed the field static chamber method to measure the flux of the hydrogen sulfide emissions during landfill operations. The CALPUFF model was utilized to simulate the diffusion of the hydrogen sulfide into the landfill and the surrounding area according to the measured flux of the hydrogen sulfide emissions. The simulation results were compared with the measured values at the monitoring point of hydrogen sulfide concentration at the field boundary. Moreover, the effects of different wind speeds, wind directions, bare working areas of the landfill, heights of the landfill, and angles between the wind direction and the long side of the working area were simulated. The main conclusions drawn from the current work can be summarized as follows:

- 1. The concentration of the hydrogen sulfide around the landfill exceeded the olfactory threshold and ranged from 0 to $60 \ \mu g/m^3$ near the boundary of the landfill. When the flux of the hydrogen sulfide emissions in the working area measured by the static chamber method was input to the CALPUFF model as the initial intensity of hydrogen sulfide pollution emission source, the simulation data was consistent with the measured results, indicating that this model was suitable for the analysis of the diffusion of the hydrogen sulfide.
- 2. The diffusion range of the hydrogen sulfide was largely on the downwind side under the influence of the wind, while the upwind direction was less affected. At a relatively uniform wind direction, the influence range of the hydrogen sulfide was limited, but it widened significantly as the wind direction changed remarkably.
- 3. At a similar bare working area of the landfill, wind speed, and wind direction, enlarging the height of the MSW dump did not considerably increase the diffusion area of

the hydrogen sulfide. Moreover, the concentration of the hydrogen sulfide declined gradually in the central area of the working area but rose in the surrounding area.

4. When the angle between the long side of the working area and the wind direction was 0°, the hydrogen sulfide largely diffused along the extension cord of the long side of the working area. Additionally, the influence range of the hydrogen sulfide noticeably extended, especially to the areas on both sides of the working area, when the angle increased to 90°.

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