Source Fingerprints of Volatile Organic Compounds Emitted from A Municipal Solid Waste Incineration Power Plant in Guangzhou, China

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Abstract

Air samples of a municipal solid waste power plant were collected for the investigation from chimney, discharge workshop, boundary site and downwind receptor sites during regular operational period as well as overhaul period. In the regular operational period, 29 VOC species were detected from the discharge workshop and 25 VOC species from the plume of chimney. Average total VOC concentration at the discharge workshop site was $57.70\pm19.55\mu g/m^3$ which was primarily composed of aromatic compounds and alkanes, but it was up to $270.63\pm2.84\mu g/m^3$ in the plume of chimney which mainly included aromatic compounds and halohydrocarbons. In the overhaul period, average total VOC concentration was $59.70\pm0.91\mu g/m^3$ in the discharge workshop and $326.35\pm151.68\mu g/m^3$ in the chimney. Aromatics, halohydrocarbons and alkanes were identified as the significant VOC fingerprints among which chlorobenzene and tetrachloroethylene were two unique VOC molecular markers of the power plant. Moreover, the VOC emission inventory of Chinese MSWI power plant was estimated approximately, it indicated that annual emission amount of the VOCs from the nationwide MSWI power plants reached 38.6tons in 2005 and would rise to 81.80 tons in the end of 2010.

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1. Introduction

With the development of intensive urbanization in the world, municipal solid waste incineration (MSWI) power plant is considered to be the most effective approaches for the disposal of municipal solid wastes by the way of minimizing waste volume and maximizing energy regeneration. Also, it
brought many disputes on how to exactly revaluate its potential adverse effects on atmospheric environment and human health\[^1\]. Therefore the emission inventory of toxic organic pollutants from these MSWI power plants was greatly underestimated or missed, such as polycyclic aromatic hydrocarbons (PAHs) and volatile organic compounds (VOCs) \[^2\].

Presently, it is widely thought that modern incineration technologies can achieve a rather high removal efficiency on the VOC up to 99.99%, but uneven temperature and incomplete combustion may still lead to the serious release or the leakage of VOCs from MSWI power plants. It is gradually proven that a long-term exposure in the polluted ambient air of VOCs might lead adverse effects on human health\[^3\]. Furthermore, VOCs are undoubtedly significant precursors to form ozone and produce secondary organic aerosols and haze\[^4,5\]. A great number of studies have been carried out to explore the emission characteristics of MSWI power plants\[^6,7\]. However, few studies have been conducted on the source fingerprinting of VOCs emitted from MSWI power plant due to the difficulty in field sampling and measuring.

Guangzhou is the capital city of Guangdong province and the primary megacity in the Pearl River Delta area (PRD) of southern China, with a population of 10.05 million in 2007. A large amount of municipal garbage resulting from fast urbanization and high density population in the city has made MSWI power plants increasingly important in PRD. Therefore, it is rather impending to identify the source emission characteristics of the MSWI power plant in PRD so that the potential risks on atmospheric environment and human health could be evaluated and reasonable mitigation measures may be taken in time.

As a typical MSWI power plant in the PRD, Likeng MSWI power plant is located in the north of Guangzhou. Since its startup in 2006, it has been subject to frequent complaints by the residents near the power plant for nuisance and potential negative health effects. This study investigated the VOC composition, the concentration and the distribution in order to well understand the VOC fingerprints and the emission characteristics of Likeng power plant. Besides, based on the estimation of its emission inventory, the potential adverse impacts on atmospheric environment and human health were discussed.

2. Experimental methods

2.1. Sampling sites

The Likeng MSWI power plant described in this study is located about 35km away from the centre of Guangzhou city that the outline of disposal process. A top level device of selective non-catalytic reduction in the current China was equipped for the disposal of flue gas, which had a maximum capacity of 1040 tons per day. Since the power plant receives the special municipal solid wastes from four districts of Guangzhou city that have a high calorific value over 5500kJ/kg, it makes the incineration primarily depending on the garbage without any additional fuels. So, the VOC components are absolutely derived from the garbage itself and its degradation.

Chimney samples were collected at site A from one small sampling port in the middle of the chimney that was 40 meters high above ground, and discharge workshop samples were taken at site B from 3m away of the garbage pit in order to escape from transferring trucks for security. Ambient air samples were taken from the boundary at site C1 and two downwind residential receptors at site C2 and site C3 which were respectively about 200m, 800m, 1000m away from the stack of the power plant. The site C1 was set on open ground, the site C2 was on the top of a three-storey residential building and the site C3 was on the roof of a four-storey building of local technical college.

2.2. Sampling methods

The sampling was performed respectively in an idle overhaul day on December 2nd and in a regular work day on December 17\(^{th}\), 2008.
In the regular operational condition, the samples from the chimney and the discharge workshop were collected at 9:00am, 10:00am and 03:00pm while the ambient samples from the receptors were collected at 9:00am, 12:00am, 03:00pm. An additional 10% samples in the amount of the total samples were duplicately conducted for quality control and quality assurance. When sampling was taken at the discharge workshop site and the ambient sites, air sample was pumped into a U-form adsorbent tube by a double diaphragm pump controlling air sampler (TH-110B, Wuhan, China) where VOCs in the air were captured by the adsorbent tube. The adsorbent tube was 4mm in diameter and 10cm in length which packed with the activated carbon in size of 100 mesh/cm² (Kechuang co., ltd, Shanghai, China). A constant flux of 0.3L/min was used to collect each air sample for 60 mins so that the sampling volume was 18L. In the chimney, the absorbent tube was conducted with an intelligent smoke-sampling instruments (TH-600C, Wuhan, China) to stretch into the sampling port of the chimney. However, the chimney samples were only collected for 10min in a flux of 0.5L min⁻¹ so that the sampling volume got to 5L.

In the idle condition for annual overhaul on December 2nd, the furnace of the power plant has been completely shut down for three days. The samples of the chimney and the discharge workshop were taken at 10:00am, 11:00am and 03:00pm while the samples of the boundary site and the ambient receptors were collected at 10:00, 14:00and 16:00. The sampling methods were the same as above in the regular operational condition.

2.3. Sample analysis

By applying an adsorption-thermal desorption unit (HL-800, Kechuang co., ltd, made in Shanghai, China), the VOC species in the sampling tubes were desorbed at 350°C for 5min into a cooling absorptive TenaxGR trap tube under a helium flow of 0.2L/min. Temperature of the TenaxGR trap tube was cooled at -10°C in order to make the pre-concentration of VOCs more effective. Then the temperature of TenaxGR tube was ramped to 300°C by 30°C /second so that the trapped VOCs were rapidly desorbed into a capillary column AT.SE-54 (60m×0.25mm×0.5m) of gas chromatograph coupled with mass detector (GC/MS, Shimadzu QP2010). The ramping program of temperature in the GC was followed as: initiated temperature at -20°C, held for 3min, rosed to 40°C by 10°C /min, then went on to 250°C by 6°C /min and held for 5min. The mass spectral detector was conducted in electron impact (EI) at 70eV ionization energy by a full scan mode in the range of 35–350 m/z. The certified VOC standards gases were purchased from USA SG Company in order to identity and quantify the VOC targets. The detection limits of all VOC targets were checked within 0.1 μg/ m³.

2.4. Quality control and quality assurance

A batch of absorbent tubes was used to collect the air samples for this study. Prior to use, all the tubes were baked up to 350°C for 3 cleaning circles under the pure nitrogen washing flow of 10ml/min and each cleaning circle was running for 5 min. The sample instrument was calibrated in field to ensure the standard deviation of sampling flow within 5%. All the collected samples were put into an amber glass jar and kept in a fridge at -4°C till analyzed by the GC/MS within 3 days. All the samples including field samples, blank samples and duplicate samples were analyzed by the identical measurement method.

3. Results and discussion

3.1. The VOC concentration level at emission source

1) The VOC concentration level in regular operation period

In the regular operation period, average total VOC concentration was 57.70±19.55μg/m³ with the range of 40.89-77.28μg/ m³ in the discharge workshop. While in the chimney, average total VOC
concentration was 270.63±2.84 g/m³ with the range of 268.62-272.64 g/m³. There were 29 kinds of VOC species detected in the discharge workshop that included 7 aromatic compounds, 2 oxygen-containing organic compounds, 17 alkanes and 2 halocarbons and one olefin. Meanwhile, there were 25 kinds of VOCs detected in chimney that included 7 aromatic compounds, 4 oxygen-containing organic compounds, 8 alkanes, 5 halocarbons and 1 olefin. During the incineration process with a high temperature of 1000°C, most of organic matters in the garbage were degraded into VOCs, CO and CO₂, and then released from the stack. Comparing with the discharge workshop, the number of detectable VOC species in the chimney was reduced but the total VOC concentration increased greatly.

It was noted that aromatic compounds were the dominant VOCs from the stack exhaust which concentration was 172.28±8.13 μg/m³ accounting for 63.66% of the total VOCs. Benzene may be formed by the recombination of C₃H₃ and the reaction of C₃H₃ and other species during burning process[8], while toluene was one of common products from garbage combustion[6,9]. However, the exhausts from transferring trucks in the power plant may also bring some influence on the volatile aromatic components. The concentration of chlorinated VOCs in chimney was 31.09±2.99 μg/m³. It increased by nearly 15 times comparing with the discharge workshop in 2.15±1.66 μg/m³, which might reason from the high concentration of hydrochloric acid (HCl) in the incinerating ash that promoted the formation of chloroethene, trichloroethylene, tetrachloroethene, as well as chlorobenzene[10]. Alkanes and oxygen-containing compounds also remarkably increased after burning. The concentration of alkanes was 15.60±4.47 μg/m³ in the discharge workshop and 43.77±4.10 μg/m³ in the chimney, and oxygen-containing compounds was 1.09±1.00 in the discharge workshop and 8.36±3.81 μg/m³ in the chimney respectively.

The concentration of olefins was relatively low at all the sampling sites but the chimney. Limonene was the most abundant olefin species with a concentration of 2.27±1.80 μg/m³.

2) The VOC concentration level in idle overhaul period

In the idle overhaul period, the total average VOC concentration of the chimney was 326.35±151.68 μg/m³ with the range of 219.10-433.60 μg/m³. It had distinctive different VOC composition from the regular operation period. It was noticed that the VOC concentration of the chimney was still higher than the discharge workshop although both furnace and ventilation system were switched off. The possible result was that the released VOCs from the discharge workshop could be concentrated in the furnace and the chimney without incineration. The total average concentration in the discharge workshop was 59.70±0.91 μg/m³ and ranged from 59.06 to 60.35 μg/m³.

Both alkanes and aromatics were the dominant VOC species in the chimney which concentrations were 158.82±56.64 μg/m³ and 159.05±89.98 μg/m³ respectively. However, they were only 10.1 and 0.8 μg/m³ respectively in the discharge workshop. The concentration of halohydrocarbons was also high up to 4.38±4.59 μg/m³ in the chimney. The total concentration of oxygen-containing compounds was 18.42±0.76 μg/m³ in the discharge workshop, which almost equaled to 1/10 of that in the chimney.

3.2. The VOC concentration level at ambient receptor sites

The VOC concentrations at the downwind ambient receptor sites of the power plant in regular operation period were higher than in overhaul period. In the regular operation, the average concentrations at site C1, site C2 and site C3 were 54.72±11.48, 77.84±10.00 and 86.89±12.07 μg/m³ respectively, but by contrast with the overhaul period, they were 29.69±5.48, 55.96±5.00 and 65.94±29.15 μg/m³ in the same sequence. It was apparent that the power plant has brought great impacts on these ambient receptor sites.

Both aromatic compounds and alkanes were the most abundant species in the two running conditions but slightly higher in the regular operational period. The concentrations of aromatic compounds were 34.93±7.34, 49.05±6.06 and 52.54±6.44 μg/m³ at site C1, site C2 and site C3 respectively in the regular operation, and 19.67±3.09, 24.61±3.66 and 26.26±7.13 μg/m³ respectively in the overhaul condition. Generally, it accounted for around 60% of the total VOCs in all sites in the regular operational period but around 50% in the overhaul condition. The concentrations of alkanes were 6.8±0.81, 19.73±4.40 and
23.00±12.78µg/m³ respectively in the regular operation, and 8.71±2.75, 21.55±3.56 and 24.89±6.52µg/m³ at site C1, site C2 and site C3 respectively in the overhaul condition. It should be noted that some aromatics and alkanes in ambient air may be contributed by the vehicle exhaust near the power plant, including benzene, toluene, ethylbenzene, 2-methylpentane and 3-methylpentane et al.

The concentrations of halohydrocarbons at sites C1 and site C2 in the normal operation period were obviously higher than in the overhaul operation period. However, a high concentration of trichloromethane with 3.71±5.79 µg/m³ detected at the receptor C3 during the overhaul period should be an unexpected occurrence and unrelated to the emissions of the power plant.

3.3. VOC fingerprint analysis

1) The VOC concentration level in regular operation period

In the two different operation periods, the concentration of specified VOC category in every sampling site was divided by its total concentration of all sampling sites in order to achieve the concentration apportionment of VOC at each sampling site.

The percentage of aromatic compounds had similar distribution characteristic between the two different operational conditions. It was mostly attributed to the plume of chimney respectively in an apportionment percentage of 49.69% in the regular operation period and 61.70% in the overhaul period, but only 10.93% and 10.94% respectively in the discharge workshop. In the regular operation condition, aromatics were made from high temperature burning of garbage in furnace and dispersed with the plume of chimney from 40m height above the ground to the downwind sites, but only released directly from the solid waste in the discharge workshop and from the closed furnace in the overhaul operation condition. So, the average apportionment of aromatics from the plume to site C2 and site C3 was 15% in the regular operation condition and dropped to 10% in the overhaul condition. Meanwhile, the average apportionment of aromatics to the boundary site C1 was changed from 10.07% in the regular operation condition to 7.63% in the overhaul condition. The apportionment of aromatics to the ambient sites was profoundly decreased during overhaul period that meant the distribution of aromatics was significantly depended on the operational condition of the power plant. Therefore, aromatics were still inferred to be one of the representative VOC fingerprint species from the chimney although they might be impacted by the local vehicle exhaust.

The percentage distribution of halohydrocarbons was drastically variable in the two different operation conditions. Halohydrocarbons coming from the process of incineration in the regular operation condition were exclusively emitted out through the plume of chimney by the apportionment of 81.51% but only 32.21% in the overhaul operation condition. However, the discharge workshop provided approximately apportionments of halohydrocarbons to the ambient sites by 5.64% in the regular operation condition and by 5.88% in the overhaul condition respectively. It caused that the concentration range of halohydrocarbons at all the ambient sites in the regular operation period was 0.14-26.70µg/m³ which was higher than in the overhaul operation period. Hence, that is why the halohydrocarbons were considered to be the second representative VOC fingerprint species from the chimney.

Alkanes from the chimney had an apportionment of 72.20% in the overhaul condition but only 38.22% in regular operation condition. The concentration of alkanes was rather approximate among the three ambient receptor sites but the apportionment was higher in the regular operation condition than in the overhaul condition. It means that the alkanes can be the third representative of VOC fingerprint species from the chimney.

Oxygen-containing organic compounds in regular operational period were mostly attributed to the chimney emission with the apportionment of 46.94%, but they were mostly attributed to the discharge workshop in the overhaul period with apportionment of 42.41%. However, it was found that the percentage of oxygen-containing organic compounds at the ambient receptor sites in the overhaul period was higher than in the regular operation period. Olefins were detected only in the source emission area of the power plant. Therefore, although oxygen-containing organic compounds
and olefins were characteristic air pollutants of the power plants, they didn’t act as appropriate VOC fingerprint species for source trace in the ambient receptor sites.

Based on the above analysis, it can be summarized that aromatic compounds, halohydrocarbons and alkanes were characteristic VOC fingerprint of the power plant.

2) Unique VOC molecular markers

Although benzene, toluene, ethyl-benzene and xylene (BTEX) were commonly considered as indicators of waste incineration, motor vehicle emissions beside the power plant.

![Figure 1. Distribution of VOC composition in different operation condition](image)

Figure 1. Distribution of VOC composition in different operation condition

also had much contribution of BTEX on the ambient air. Hence, we focused on the independent and unique VOC molecular markers from the power plant.

a) Chlorobenzene

Chlorobenzene widely occurs in the disposal processes of municipal solid waste[6,10-12]. The distribution of chlorobenzene at all our sampling sites was shown as in Fig. 3. Average concentration of chlorobenzene in the regular operational chimney was 8.29±2.67 μg/m³, which a comparable research result was 2.11 μg/m³ in one German MSWI studied by Jay[11]. Noticeably, the concentrations of chlorobenzene in discharge workshop and in receptors sites during the regular operation period were too low in the range of 0.06-0.10 μg/m³ to be detectable at the ambient sites in the overhaul period. However, due to the rather stable and strong benzene ring structure, chlorobenzene has a long atmospheric lifetime of about 13-day before it is removed by reaction with OH[13]. Therefore, chlorobenzene was one of significant unique molecular markers for the MSWI power plant.

b) Tetrachloroethylene

Tetrachloroethylene is usually associated with dry cleaning and metal cleaning agent[14] and is also abundant in open cells receiving municipal solid waste[15]. Old clothes soaked with tetrachloroethylene and trichloroethylene were sometimes found in the waste[16]. Average concentration of tetrachloroethylene in the regular operational chimney was 1.42±0.31 μg/m³, which a comparable result was 0.16 μg/m³ in the German MSWI studied by Jay[11]. Although tetrachloroethylene showed low but rather stable concentration in the range of 0.46-2.57 μg/m³ in all receptor sites, it was undetected in the discharge workshop.

Tetrachloroethylene also has a rather long atmospheric lifetime of about 0.4 year before it is removed by reaction with OH[17]. In the overhaul period, tetrachloroethylene was often below the detection limit. Moreover, there were no more primary emission sources of tetrachloroethylene such as dry cleaning and metal cleaning in the investigated area. Hence, tetrachloroethylene was the other important unique molecular markers of the power plant, which was consistent with the found of Chiriac[18].
Although all the VOC targets could be explored to construct the source profiles of the MSWI power plant, the VOC species with a long photochemical lifetime and low activity in airborne reactions would be the better representative of source profile. Based on this consideration to reconcile the source profile, there were 20 VOC species selected to conduct a normalized calculation. The individual VOC concentrations were divided by total concentration of the selected VOCs, therefore the attribution of each species was acquired. Also the standard deviations for each species were calculated and shows as Figure 4.

This normalization may not only avoid the confusion caused from a widely range of measured VOC concentrations but also well describe the emission characteristics in a complex dynamic conditions. It was supposed to provide scientific basis for source identification, emission inventory and pollution control.

It is clear that the seven VOC species of No.1, 2, 3, 6, 7, 10 and 17 in the source profile for the chimney accounted for considerable apportionments. Chlorobenzene (No.15) and tetrachloroethylene (No.16) were two remarkable molecular markers of the source emission, which were important reactants in atmospheric photochemical reactions and promoted the production of gaseous halogen precursors such as CINO2 and CIONO2 when reacting with NOx under similar pressure and thermal condition of flue gas emission[19]. Cai et al. demonstrated that airborne radical C1 can oxidize toluene, which plays an role in the SOA production as important as OH[20]. Profound differences of the source profile between the regular operational and the overhaul period were observed not only the species but also the quantities due to the incineration which a critical process for oxidation and thermal decomposition and thus for the genesis of many PAHs.

In the source profile of the discharge workshop, the seven VOC species of No.1, 2, 4, 6, 7, 14 and 18 achieved large apportionments. Methylyclohexane in No.19 was one of significant species in the overhaul period which had high potential to produce secondary organic aerosol than straight-chain alkanes[21]. The source profile of discharge workshop was similar in two different operational conditions. The apportionments of aromatics and halocarbons in the regular operation period were remarkable larger than in the overhaul period.
3.4. Discussion on the potential negative impacts

Many VOCs released from the power plant may either impair the sensory system or the respiratory system of human body. Some can cause carcinogenic or mutagenic impacts on human after chronic exposure. Bird et al. found that benzene was able to induce aplastic anemia, bone marrow, myeloblastic anemia and genotoxic effects in humans at trace level\cite{22}. Styrene and chlorobenzene could induce the production of inflammatory mediators in lung cells\cite{3,23}. Therefore, it is significantly noticeable that the five carcinogens and possible carcinogens VOC species identified by IARC\cite{23} were detected in this study which included benzene, ethylbenzene, styrene, trichloromethane, and tetrachloroethylene.

The flux of the chimney in Likeng MSWI power plant was currently about 243,000 m$^3$/h\cite{24}, which means that the annual equivalent emissions of total VOC from the chimney got to 576.08kg under a continuous 24-hour regular operational condition. By a nationwide statistical investigation in 2005, there have already existed 67 MSWI power plants throughout china. Supposing all these plants were equipped with the combustion and flue gas treatment devices as excellent level as that in Likeng MSWI power plant, and the VOC concentration and the total VOC emissions amount were equalled to the Likeng MSWI power plant, then we may make a rather rough but conservative estimation for the VOC emission inventory in China. Therefore, the annual emission amount of total VOC from the current nationwide MSW incineration power plants is at least 38.6 tons.

According to the primary schedule in the Chinese 11th Five-Year developing Plan, there will build 75 new MSWI power plants during 2006-2010. If based on the same calculation method as the above, the nationwide emission of total VOCs from the MSWI power plants in China will increase to 81.80 tons annually, which means that this emission source must pose a potentially significant threats to urban air quality and human health in the very soon future. Unfortunately, there are still no strict emission controlling limitations for the VOC concentration and the total amount included in the present Chinese Solid Waste Incineration Pollution Control Standard(GWKB3-2000) which was promulgated in 2000. Therefore, it suggested that a definitely emission limitation for individual VOC and total VOCs should be made through this study. It is possibly that the current air pollution emission standard regulated in Europe would provide useful help to develop Chinese national standard for the total VOCs from MSW incineration power plants.

4. Conclusion

This study explored the VOC composition, the molecular markers and the source profile for the MSWI power plant in Pearl River Delta area, China. Under the regular operational condition, there were 29 VOC species detected in the discharge workshop with the total VOC concentration of 57.70±19.55µg/m$^3$, while there were 25 VOC species detected from the chimney with the total VOC concentration of 270.63±2.84µg/m$^3$. In the overhaul period, the total VOC concentrations were 59.70±0.91µg/m$^3$ and 326.35±151.68µg/m$^3$ respectively in the discharge workshop and the chimney.

Aromatic hydrocarbons and halohydrocarbons were inferred as the significant VOC fingerprint species of chimney emission, and some priority control strategies should be made for their reduction. Chlorobenzene and tetrachloroethylene were identified to be unique molecular markers of the Likeng MSWI power plant. The VOC fingerprints in the chimney of the MSWI power plant showed significant difference between the regular operational period and the overhaul period.

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