

# Development of evaluation method for explosives trace detection with non-contact sampling

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## Abstract

Explosives trace detection (ETD) characterized by non-contact sampling is a promising technology to enable high-throughput and automatic detection. In this study, we attempted to develop a standard sample and evaluation method for ETD. Soil particulate was coated with trinitrotoluene (TNT) to be used as a standard sample. Adsorption of TNT was confirmed by liquid chromatography-mass spectrometry. The standard sample was filled in the hollow on a metal plate that was used as the test target. The hollow plays a role for maintaining the amount and the position of the sample between measurements. We obtained the sensitivity of ETD using the standard sample and test target. Collection efficiency, a pure performance of the sampling part of the ETD, was also acquired. The standard sample and evaluation method using that sample and the plate with the hollow will be useful for the improvement of ETD with non-contact sampling.

**Keywords:** explosives trace detection, mass spectrometry, particulate, non-contact sampling, evaluation standards

## 1. Introduction

The threats of terrorism have been a serious problem around the world. More than 10,000 terrorism acts occurred in 2015, resulting in the deaths of approximately 30,000 people<sup>1</sup>. Because explosives are used for more than 50% of terrorism acts<sup>1</sup>, the technology to detect explosives is of high importance as counterterrorism. To find hidden explosives, there are two types of explosives detectors: bulk detection and trace detection<sup>2,3</sup>. Bulk detection methods include X-ray imaging, millimeter wave imaging, nuclear quadrupole resonance, etc. that are used to determine the shape and size of concealed objects in a covered material such as luggage<sup>4</sup>. Bulk detection cannot, however, easily identify what the concealed objects are.

Trace detection, on the other hand, which includes chemical analysis methods based on mass spectrometry (MS), ion-mobility spectrometry (IMS) and chemiluminescence, has been applied to detect the existence of trace contaminants of explosives adhered to a passenger's body, clothes, and luggage<sup>5</sup>. IMS with swab sampling is widely accepted as a gold standard for trace explosives detection<sup>6</sup>. MS is a powerful analytical method characterized by high sensitivity and high resolution. Such characteristics lead to a low false positive rate, which has been confirmed in a filed test<sup>7</sup>. MS is therefore an alternative to IMS<sup>8-14</sup>. Trace detection identifies explosives, and its selectivity is higher than that of bulk detection. It cannot, however, determine the amount of

explosives in a suspicious item such as luggage. As mentioned above, the characteristics of bulk detection and trace detection are different. The combined use of bulk detection and trace detection therefore improves the possibility of finding hidden explosives.

At current security checkpoints in airports, not all passengers and carry-on luggage are inspected by trace detectors, although all luggage is examined by X-ray imaging. One of the reasons is that the throughput of the trace detectors is insufficient. To overcome the problem concerning throughput, we previously developed a high-throughput ETD, which is characterized by non-contact sampling<sup>15,16</sup>. The ETD we developed enables the detection of explosives within a few seconds. Such high throughput detection can lead to the realization of inspection of all passengers.

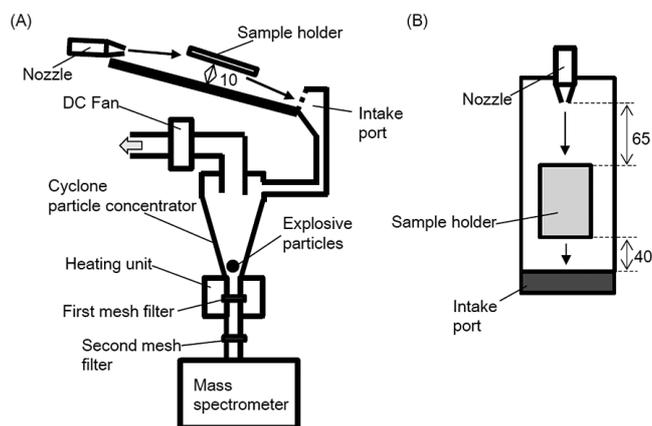
As ETD is security-related equipment, its performance is certified by authorities. The Transportation Security Administration (TSA) and the European Civil Aviation Conference (ECAC) have certified the ETD with swab sampling, although standard test sample and evaluation method have not been disclosed. On the other hand, the American Society for Testing and Materials (ASTM) international published the evaluation method and 8 groups of explosives that should be detected<sup>17</sup>. The concept of such a method is that the mixture of the explosive sample and defined background material is used as a standard sample. In addition, dry transfer samples, one of the standard explosive samples for the evaluation of those detectors, is also open to the public in the form of a patent<sup>18</sup>. Moreover, National Institute of Science and Technology provides micro particle of silica coated with explosives as a calibration sample<sup>19,20</sup>. Those samples and methods are geared towards ETD with swab sampling.

ETD characterized by non-contact sampling of explosive particulate is an emerging technology to enable high-throughput and automatic detection. Not only us but also some companies and research groups developed non-contact sampling ETD. However, different from ETD with swab sampling, there is no standard test sample and evaluation method for ETD with non-contact sampling. In this study, therefore, in reference to the ASTM international, we attempted to establish that sample and method and to evaluate sensitivity and collection efficiency of our ETD.

## 2. Experimental

### 2.1 Experimental setup

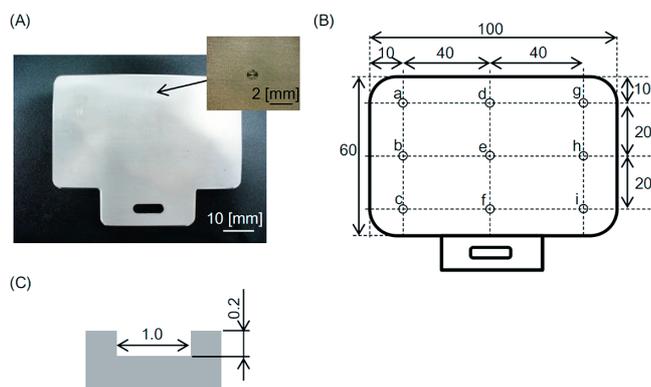
A schematic diagram of the non-contact sampling ETD which we developed is shown in Figure 1(A). A nozzle, which is connected to an air compressor (not shown in the figure), emits compressed-air jets to a sample holder where the explosive particulates are attached. An intake port is also located at the opposite side of the nozzle to collect detached particulates. The inner diameter of the nozzle is 1 mm, and the pressure of the compressed air was set at 0.1 MPa. The number of air jet emissions was set to three times per test. The duration time of the jet was set to 0.1 s with intervals of 0.1 s. The particulates



**Figure 1** Experimental setup. (A) Explosives trace detection system with non-contact sampling. (B) Positional relationship between a sample holder, a nozzle and an intake port.

detached from the sample holder are introduced into a cyclone preconcentrator via the intake port<sup>15</sup>. The flow rate at the cyclone was set at approximately 40 L·min<sup>-1</sup>. The particulate is separated from the air flow by the cyclone preconcentrator and deposited on the first filter (GF-D1N, NIPPON SEISEN, Co., Ltd.). The particles are vaporized by heat, and the vaporized molecules are introduced into the ion source of an ion-trap mass spectrometer via a second filter (GF-D1N, NIPPON SEISEN, Co., Ltd.). The temperature of the vaporizer, the transfer tube that connects the vaporizer and the ion source, and the ion source was set to 180°C. The flow rate of the sample gas from the cyclone preconcentrator to the ion source was set to 0.5 L·min<sup>-1</sup>. The vaporized molecules are then ionized by an atmospheric-pressure chemical ionization with counter-flow introduction (APCI-CFI)<sup>21</sup>. To detect 2,4,6-trinitrotoluene (TNT), a negative ionization mode was used. The ions produced by the APCI-CFI are analyzed by the ion trap mass analyzer<sup>22)–25)</sup>. The ion-accumulation time and the scanning time were set at 50 ms and 100 ms, respectively.

Figure 2(A) is a photograph of the sample holder with hollows which is made of stainless. Its thickness is 6 mm. The hollows on the holder are filled with a standard sample, which is particulate coated with TNT as described below. The hollow plays a role for maintaining the amount of the standard sample for each measurement. Three sizes of the hollow are prepared:  $\phi$ 1.5 mm, 0.5 mm depth,  $\phi$ 1.0 mm, 0.5 mm depth and  $\phi$ 1.0 mm, 0.2 mm depth. The location of the hollows and a section of one of the hollows are illustrated in Figure 2(B), (C). The positional relationship between the sample holder and nozzle is also illustrated in Figure 1. The sample holder was placed in parallel with the sampling surface with a 10 mm space. In addition, the sample holder was separated from the nozzle and the intake port by 70 mm and 40 mm, respectively. The nozzle, the center of the sample holder and the intake port were aligned. The surface of the sample holder where the hollows are is set downward, and the side of the hollow a, b and c is set close to the side of the nozzle. In the measurement, the standard sample filled in the hollow is



**Figure 2** Sample holder. (A) Photographs of a sample holder with a hollow. (B) Position of hollows dug on the surface of the metal plate. (C) Section of a hollow.

detached by the air jet and collected by the intake port.

## 2.2 Reagents

The solution containing TNT was purchased from AccuStandard Inc., USA. It was diluted with methanol to obtain suitable concentrations. Japan Industrial Standard (JIS) Z 8901 Kanto-loam powder (fine particle), which is soil from the Tokyo area of Japan (hereafter called “Tokyo soil particulate”), was used as the particulate to be coated with TNT. The diameter of approximately 80% of the particulate ranged from 5 to 75  $\mu\text{m}$ . Tokyo soil particulate was also used as a background material.

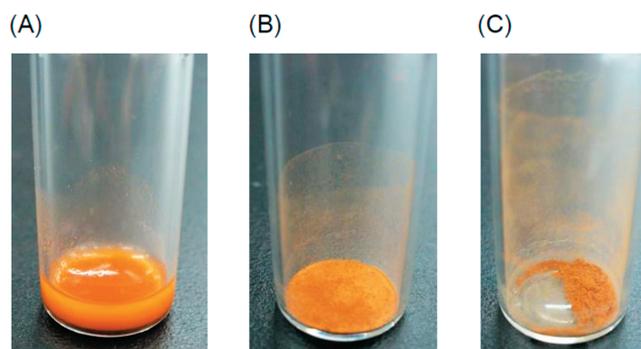
## 2.3 Preparation of standard sample

Tokyo soil particulate of 20 mg was prepared in a 3-mL vial using a medicine spoon. Methanolic TNT at a proper concentration was mixed with the particulate (Figure 3 (A)) and dried at room temperature. The dried particulate was resuspended with 300  $\mu\text{L}$  of acetone and dried at room temperature (Figure 3(B)). Afterward, the dried particulate was powdered using a fine stick (Figure 3(C)). Prepared particulate is defined as “standard sample.” Until the measurement, the standard sample was stored at  $-30^\circ\text{C}$ . For the measurement, the standard sample was placed in the hollow of the sample holder using a spatula at room temperature.

## 2.4 Confirmation of prepared standard sample

The amount of the standard sample in the hollow of the sample holder was estimated by measuring the weight of the standard sample using a scale (AUW120D, Shimadzu). The standard sample containing 0.03% TNT was placed in the hollow of the sample holder. This sample was collected in a 1.5 mL tube, and the same process was repeated five times. The weight difference of the tube before and after the collection was measured and divided by 5 to calculate the average weight of the standard sample filled in the hollow for one time. This experiment was only conducted using the hollow with  $\phi 1.5$  mm, 0.5 mm depth and not using smaller hollows because the measurement of the weight of the smaller amount of the standard sample was unstable.

In addition to the weight measurement, the amount of



**Figure 3** Photographs of the process of sample preparation. (A) Mixture of particulate and a solution containing TNT. (B) Dried sample coated with TNT. (C) Standard sample reduced to a powder.

TNT in the hollows was quantified by mass spectrometry. The standard sample containing 0.03% TNT was filled in the hollow of the sample holder. This sample was collected in a 1.5 mL tube. Then, 1 mL of methanol was added and mixed using a vortex mixer for 1 min. The concentration of TNT in that solution was quantified by liquid chromatography-mass spectrometry (LC-MS, Q Exactive, Thermo Fisher Scientific Inc.). Water with 1% acetic acid and methanol with 1% acetic acid were used for the mobile phase. A C18 LC column was used (InertSustain C18, GL Sciences Inc.).

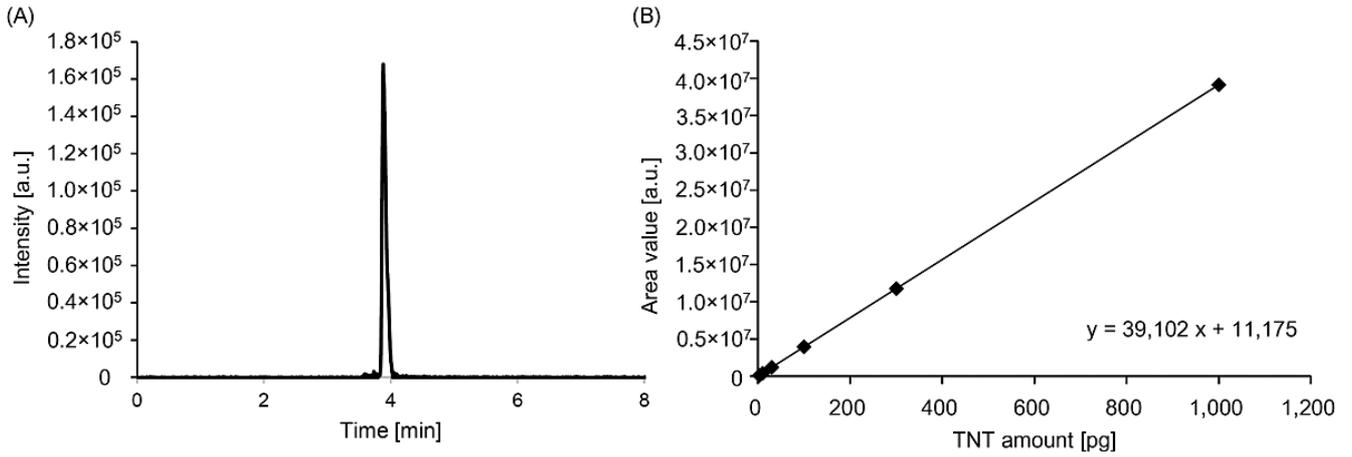
## 2.5 Measurement of standard sample by explosives trace detection system

To obtain a calibration curve for TNT, the standard sample containing TNT at a proper concentration was filled in the hollow of the sample holder. The position of the hollow was as illustrated in Figure 2. The sample holder with the standard sample was then set at the explosives trace detection system. The positional relationship between the sample holder and the system is shown in Figure 1. An extracted ion chromatograph (EIC) for  $m/z$  226 ion, which is  $[\text{M-H}]^-$  ion of TNT, and its peak area were obtained. The photos of the standard sample in the hollow were obtained using a USB microscope (Dino-Lite Premier, Thanko, Ltd.) before and after the measurement. The sample holder having the hollow with  $\phi 1.0$  mm, 0.2 mm depth was used for this experiment.

Collection efficiency of the ETD was defined by the ratio of the amount of TNT molecule introduced into mass spectrometer to that on the sample holder. To calculate such efficiency, we built a calibration curve showing the relationship between the TNT amount introduced into the mass spectrometer and the peak area obtained in the EIC by a liquid injection method, where the solution containing TNT at the proper concentration was dropped on the first mesh filter. The standard sample containing 0.1% TNT was filled in 9 hollows of the sample holder individually, and the peak area for TNT in the EIC was measured. The obtained peak area was converted to the collection efficiency using the calibration curve.

## 3. Results

The average weight of the standard sample filled in the



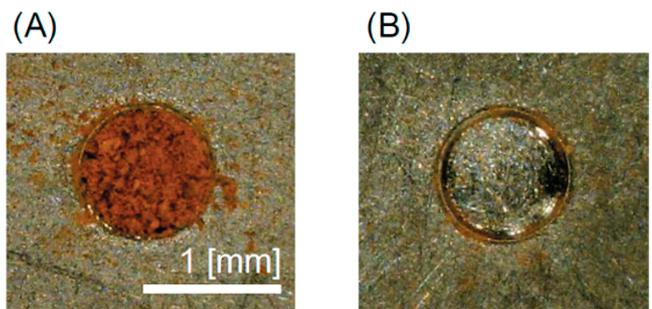
**Figure 4** Results of LC-MS. (A) Extracted ion chromatogram for TNT from standard sample containing 0.03% TNT. (B) Normalization curve for TNT used in quantification.

**Table 1** Sample amount in a hollow.

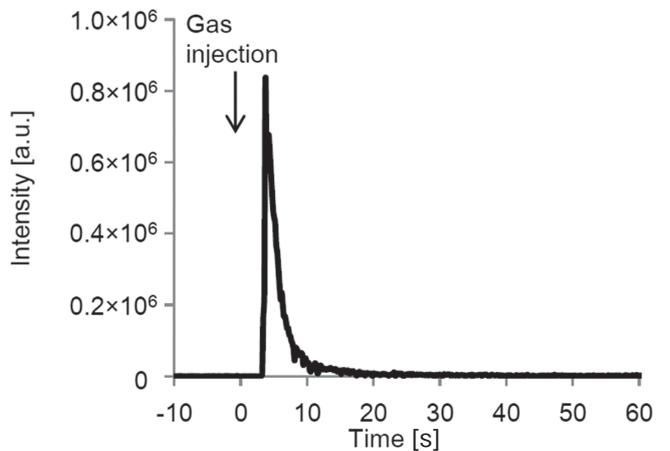
Size	$\phi$ 1.5, 0.5 [mm]	$\phi$ 1.0, 0.5 [mm]	$\phi$ 1.0, 0.2 [mm]
Quantified TNT amount [ng]	93 ± 27	37 ± 6.3	15 ± 3.2
Test sample weight [ $\mu$ g]	310 ± 89	123 ± 21	48 ± 11
Estimated TNT amount [ng]	95 ± 4.8	42 ± 2.1	17 ± 0.9
Estimated test sample weight [ $\mu$ g]	318 ± 16	141 ± 7.0	57 ± 13

hollow with  $\phi$  1.5 mm, 0.5 mm depth was measured using a weight scale. As shown in Table 1, the weight was  $310.3 \pm 88.7 \mu\text{g}$ . Based on the volume difference between the hollows, the standard sample weight filled in the hollows with  $\phi$  1.0 mm, 0.5 mm depth and that with  $\phi$  1.0 mm, 0.2 mm depth was calculated. The weight for those hollows was  $123.2 \pm 21.0 \mu\text{g}$  and  $48.3 \pm 10.8 \mu\text{g}$ , respectively (Table 1). The TNT amount recovered from the standard sample filled in the hollows was quantified by LC-MS. Figure 4 shows the representative EIC for TNT obtained using standard sample filled in the hollow with  $\phi$  1.0 mm, 0.2 mm depth and the normalization curve for TNT in LC-MS. The quantified TNT amount was  $93.1 \pm 26.6 \text{ ng}$ ,  $37.0 \pm 6.3 \text{ ng}$  and  $14.5 \pm 3.2 \text{ ng}$  for the hollows with  $\phi$  1.5 mm, 0.5 mm depth,  $\phi$  1.0 mm, 0.5 mm depth and  $\phi$  1.0 mm, 0.2 mm depth, respectively (Table 1). Using the quantified TNT amount and the concentration of TNT of the standard sample, which was 0.03% in this experiment, the standard sample weight filled in the hollow was estimated. The estimated standard sample weight was  $318 \pm 16 \mu\text{g}$ ,  $141 \pm 7.0 \text{ ng}$  and  $57 \pm 13 \mu\text{g}$  for the hollows with  $\phi$  1.5 mm, 0.5 mm depth,  $\phi$  1.0 mm, 0.5 mm depth and  $\phi$  1.0 mm, 0.2 mm depth, respectively (Table 1).

The TNT amount, which was quantified by LC-MS, was similar to the estimated TNT amount, which was calculated from the standard sample weight, suggesting that expected amount of TNT was adsorbed on the particulate. In the following experiment, when the TNT amount used for the experiment was changed, the concentration of TNT was changed with constant standard sample weight by using only the hollows with  $\phi$  1.0 mm, 0.2 mm depth. For simple calculations, the



**Figure 5** Photographs of standard samples. (A) before emission of the air jet from the nozzle, and (B) after emission of the air jet.



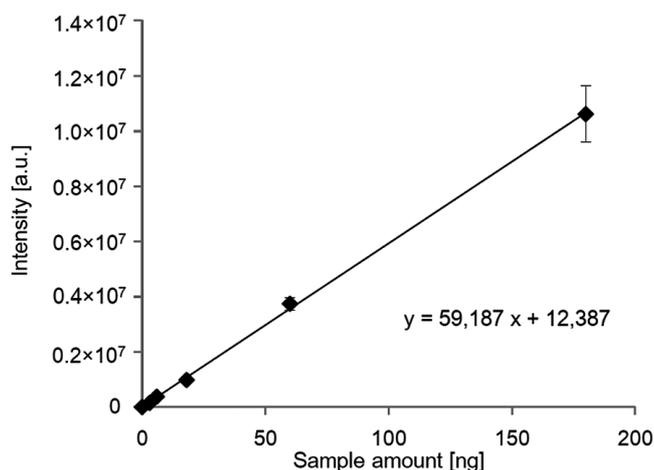
**Figure 6** Extracted ion chromatogram for TNT in the explosives trace detection system.

standard sample weight filled in that hollow was defined as  $60 \mu\text{g}$ .

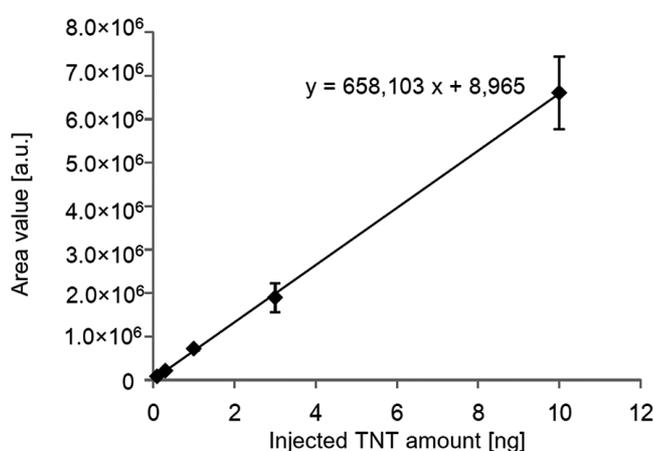
Figure 5 shows the photographs of the standard sample on the sample holder before and after the emission of the air jet.

The air jet detached the standard sample completely. The EIC obtained for TNT is shown in Figure 6. The x-axis indicates the time after the emission of the air jet. The TNT signal could be observed within a few seconds after the emission.

The EIC was recorded for TNT at several concentrations. Peak area was calculated and used for the



**Figure 7** Relationship between the TNT amount and the peak area in the standard sample measurement using the explosives trace detection system.



**Figure 8** Normalization curve for TNT in the liquid injection method.

calibration curve (Figure 7).

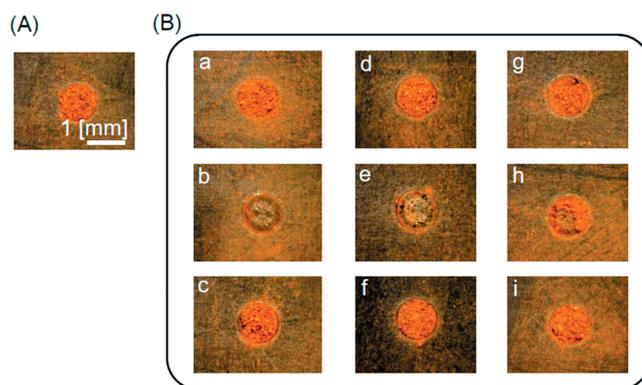
The TNT amount shown in the x-axis of Figure 7 was calculated using the concentration of TNT and the standard sample weight of 60  $\mu\text{g}$ . Figure 7 demonstrates a clear linear relationship between the TNT amount and the peak area of the EIC, suggesting that the method is useful for the evaluation of the sensitivity of the equipment. The limit of detection (*LOD*) was calculated by the following equation:

$$LOD = \frac{i + 3\sigma}{a} \quad (1)$$

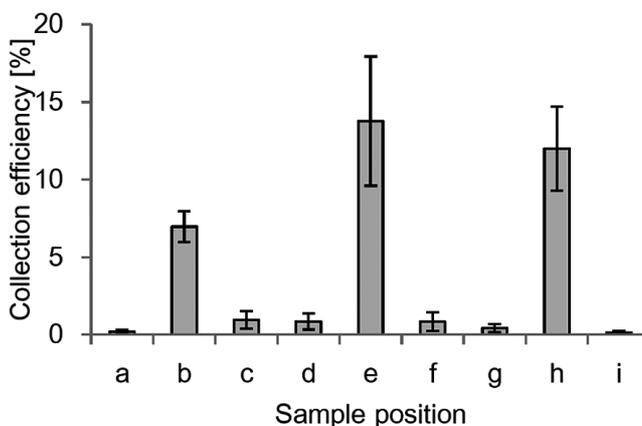
where  $i$  and  $a$  are a y-intercept of the fitted curve in Figure 7 and its slope, and  $\sigma$  is the standard deviation of a blank signal. The *LOD* was calculated to be 0.3 ng.

The relationship between the TNT amount introduced into the MS and the peak area in the EIC was recorded by a liquid injection method; the results are shown in Figure 8.

This curve was used for the calculation of the collection efficiency. The positional dependence of this efficiency was measured by filling the standard sample in all 9 hollows in the sample holder. Figure 9 shows the photographs of the standard sample in each of the hollows of the sample holder before and after the emission of the air jet.



**Figure 9** Dependency of the detachment efficiency on sample positions. (A) Photograph before emission of the air jet from nozzles. (B) Photograph after emission of the air jet. Letters indicate the sample positions and correspond to those in Figure 2.



**Figure 10** Dependency of the collection efficiency on sample positions. Letters indicating the sample positions correspond to those in Figures 2 and 8.

Only the standard sample at the positions b, e and h, which are coaxially arranged with the air nozzle, was removed. This implies that the air jet has directionality and does not reach the standard sample located away from its direction. From the peak area in the EIC and the normalization curve in Figure 8, the TNT amount arriving at the MS was calculated. That value was divided by the TNT amount on the sample holder, calculating the collection efficiency at each position of the sample holder (Figure 10).

Because the standard sample at the positions a, d, g, c, f and i was not detached, as shown in Figure 8, the collection efficiency was close to zero. On the other hand, the efficiency at positions b, e and h was approximately 10%. Because approximately 100% of the standard sample was detached at positions b and e, as shown in Figure 9, 10% collection efficiency means that 10% of the standard sample detached from the hollow and arrived at the MS.

#### 4. Discussion

In this paper, an attempt was made to prepare a standard sample and to develop an evaluation method for ETD characterized by non-contact sampling of particulate. TNT was coated on the Tokyo soil particulate, which is soil from the Tokyo area of Japan. The diameter of such soil ranges from 5 to 75  $\mu\text{m}$ . ASTM international proposes

a soil from the USA to be used for background material mixed with explosives and used for the standard sample in the evaluation of ETD<sup>13</sup>. The sensitivity under the effect of the background material can be obtained. Verkouteren et al. (2010) analyzed the diameter of explosive particulate in fingerprints and suggested that 10–20  $\mu\text{m}$  explosive particulate was a main target of ETD<sup>26</sup>. Those ideas are reflected in our preparation of the standard sample. Similar preparation was previously done for the standard sample of narcotic<sup>27</sup>. By using particulate whose diameter is adjusted, reproducible sample can be obtained.

To confirm the sample condition, the amount of TNT coated on the particulate was quantified by LC-MS. That amount was also calculated from the standard sample weight filled in the hollow of the sample holder and the TNT concentration. The TNT amount obtained from both methods was similar (Table 1), suggesting that TNT was adsorbed on the particulate as expected. The Tokyo soil particulate consists of 40%  $\text{SiO}_2$ . TNT is likely binding to the silica in this particulate.

For the measurement of the standard sample by our ETD system, we prepared a sample holder with a hollow where the standard sample is placed. That hollow maintains a standard sample amount and the positional relationship between the sample and the nozzle and the intake port for each measurement. In the performance evaluation of the ETD system, the most important evaluation item was the *LOD* for explosives, that is, the sensitivity of the equipment. The *LOD* can be obtained using a calibration curve. We measured the standard sample containing TNT at various concentrations in the condition of a constant standard sample weight (Figure 7) to acquire the sensitivity under the constant effect of background material. Figure 7 shows a clear linear relationship between the TNT amount on the sample holder and the area of the curve in the EIC, suggesting that our proposed method is useful for the determination of the sensitivity of the equipment.

The sensitivity of the equipment is the product of the sensitivity of the sensor, which is MS in this experiment, and the collection efficiency. The collection efficiency is a pure performance of the sampling. Different from the contact sampling, the collection efficiency in the non-contact sampling has a positional dependency. To evaluation such dependency, the hollow filled with the standard sample was set in several locations of the sample holder (Figure 2). As shown in Figures 9 and 10, only the standard sample coaxially arranged with the air nozzle was collected. Such positional dependency of the collection efficiency should be useful information for the improvement of the non-contact sampling system. In the contact sampling system, improvements in a swab paper, such as a change of material and a coating of swab paper, change the collection efficiency<sup>28)–30)</sup>. On the other hand, a control of the air jet, such as a change of the nozzle shape, the number of the nozzles and air pressure, improves the collection efficiency in the non-contact sampling.

ETD with non-contact sampling of particulate is an emerging technology to enable high throughput and

automatic detection. We and other several research groups have attempted to develop a non-contact sampling system<sup>31)–34)</sup>. The standard sample and evaluation method we propose here would be useful for researchers and manufacturers of such systems.

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