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A New Wipe-Sampling Instrument for Measuring the Collection Efficiency of Trace Explosives Residues

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Abstract

Trace explosives detection, a crucial component of many security screening environments, commonly employs wipe-sampling. Since collection of an explosive residue is necessary for detection, it is important to have a thorough understanding of the parameters that affect the efficiency of collection. Current wipe-sampling evaluation techniques for explosive particles have their limits: manual sampling (with fingers or a wand) is limited in its ability to isolate a single parameter and the TL-slip/peel tester is limited to a linear sample path. A new wipe-sampling instrument, utilizing a commercial off-the-shelf (COTS) 3D printer repurposed for its XYZ stage, was developed to address these limitations. This system allowed, for the first time, automated two-dimensional wipe-sampling patterns to be studied while keeping the force and speed of collection constant for the length of the sampling path. This new instrument is not only capable of investigating the same parameters as current technology (wipe materials, test surfaces, forces of collection, and linear sample patterns), it has added capabilities to investigate additional parameters such as directional wipe patterns (i.e. “L” and “U” shapes, square, and serpentine) and allowing for multiple lines to be sampled during a single collection without the need for adjustments by the user. In this work, parametric studies were completed using 1,3,5-trinitroperhydro-1,3,5-triazine (RDX) and the COTS 3D printer for wipe-sampling to establish collection efficiencies for numerous scenarios. Trace explosives detection in field screening environments could be greatly improved with the ability to comprehensively investigate how a wide range of parameters individually affect collection by wipe-sampling. A screener who knows how to properly interrogate any given surface will be much more efficient at detecting trace explosives.

Keywords

Wipe Collection; Trace Explosives; Standardized Sampling; Collection Efficiency

Introduction

The methods for interrogating a surface to detect contamination are as varied as their applications. There are direct sensing techniques such as X-ray devices for metal detection,

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radiation meters for radioactivity, and photoionization monitors for volatile organic compounds,¹ as well as indirect detection methods which require the collection of the contaminant followed by a subsequent analysis step. This type of indirect chemical screening is widely employed in the trace contamination detection applications. The process of interrogating a surface, and analyzing the particles collected is widely used in forensics,^{2–6} and environmental and regulatory screening.^{7–11} Many agencies have standardized methods for collecting a targeted contaminant and these methods can vary greatly depending on the target particles, the analysis, and the surface itself. For many of the environmental and regulatory screening applications, the focus is often on quantitative collection,^{12–18} decontamination,¹⁹ or a demonstration of cleanliness^{20,21} and therefore the collection method is focused on achieving those goals. Vacuum techniques²² may be employed when noncontact is beneficial. If physical contact with the surface is acceptable then a roller,^{23–25} tape,^{26,27} or wipe can be used. A wide range of wipe materials have been used (cotton or rayon gauze pads, microfiber paper, or filter paper)¹ and in many applications a solvent is applied to the wipe to assist in collection.^{23,24,28–31} Direct and indirect sensing techniques are currently widely used in both government and private sectors.

While there are many applications for wipe-based collection, this work is focused on a specific application—trace explosives collection in an operational setting for rapid, qualitative analysis. Trace explosives detection by wipe-sampling is widely used in the homeland security^{32–34} sector as it is commonly coupled with trace explosives detection technologies such as ion mobility spectrometry (IMS). Given that this type of analysis commonly occurs in a high-volume venue, the collection requirements are much different than common environmental applications and include high reliability, speed, and direct interaction and sampling of individuals.³⁴ A successful detection of trace explosives requires the collection of residual explosive particles from a person or their belongings after they have come in direct or secondary contact with bulk explosives. Detection can also occur by the collection of particles present in a contaminated fingerprint that has been deposited on a surface. Samples are collected by wiping a surface with a fabric wipe and introducing that wipe into a trace explosives detector which then thermally desorbs the explosive particle off of the wipe for detection. Thermal desorption is the common sample introduction technique; therefore, the wipes must be suitable to high temperatures and cannot employ a solvent to assist in collection. Research is continuing to provide a better understanding of the parameters surrounding the collection aspect to improve the likelihood of detection using wipes suitable for trace explosives detection by IMS.

Successful detection of a residue in this type of setting requires three separate steps. The first step requires the surface be contaminated with an explosive residue. Secondly, the surface must be interrogated with a collection wipe using a sampling wand or a wipe-sampling instrument. Finally, the wipe must be analyzed for the presence of the analyte through direct analysis. While significant research has focused on improvements of the detection technologies, the third step, less work has been done to better understand what is required to successfully collect a residue.

Understanding what factors affect the collection of a residue off a surface requires a reproducible method for sample deposition. For this type of work, preparation of a test

surface contaminated with an explosive analyte typically uses a dry-transfer approach.³⁵ First, a concentrated solution of the analyte is pipetted or printed onto a smooth polytetrafluoroethylene (PTFE) substrate and allowed to dry. The analyte is then transferred to the test surface by placing the PTFE substrate at the desired location, analyte side down, and translating the PTFE substrate 2 cm – 3 cm across the test surface with moderate applied pressure. This approach relies on the drying process to produce appropriate analyte particles with characteristics (such as size and shape) that simulate residues collected from fingerprint deposition after handling bulk explosives.^{36–41} Trace explosive particles transferred through fingerprint deposition, such as RDX and pentaerythritol tetranitrate (PETN) from handling C-4 and Semtex H, ranged from less than 1 μm to greater than 50 μm in diameter.⁴² Early studies^{36,43,44} have used fluorescent polystyrene latex (PSL) microspheres within that particle size range. Though only a model, the ability to sample discrete particles sizes with these microspheres allows for a better understanding of how the size of the particles will affect collection. The fluorescent property of the PSL microspheres allows for direct analysis of the wipe and test surface without the need for extraction and are therefore useful model particles. However, their dissimilarity to explosives in chemical and physical properties is a limitation. Verkouteren *et al.*⁴⁵ have addressed this limitation by preparing RDX samples through drop-on-demand (DOD) inkjet printing. This approach results in a higher level of control over the size and chemical composition of the particles used for dry-transfer.

Until recently, a standardized method for measuring the collection efficiency (CE%) of explosive particles after interrogating a test surface with a wipe had not been established. Verkouteren *et al.*⁴⁶ addressed this by describing key factors to consider when designing wipe-sampling experiments. The capabilities of the wipe-sampling instrument and features to consider when choosing wipes and test surfaces to investigate were discussed. A protocol for the preparation of analyte particle standards by DOD inkjet printing was outlined. Finally, a detailed procedure for wipe-sampling that included steps for dry-transfer, collecting the analyte onto a wipe, extracting from the wipe, and analysis was discussed. By proposing a standardized test method, the focus can now turn towards comprehensively investigating the effects of individual parameters of the wipe-sampling process: the wipe and test surfaces, the wipe-distance, the force of collection (applied load), wipe-sampling pattern, and lifting the wipe multiple times throughout a single wipe-sampling collection event. These factors are so integral to effective explosives detection by wipe-sampling that the U.S. Department of Homeland Security developed a working group⁴⁷ to better understand the role the different parameters play in the CE% of trace explosives by wipe-sampling.

Previous wipe-sampling efficiency studies^{36,46,48,49} for trace explosives detection have typically used a TL-slip/peel tester to allow for programable and reproducible speeds and wipe distances. The TL-slip/peel tester is a useful instrument; however, it has its limitations. It moves the surface beneath the wipe, not the wipe across the surface and is limited to movement in a linear direction. Another disadvantage is the requirement for the user to place the wipe holder in the appropriate location. This placement is key to the wipe traveling over the deposited analyte (roughly a 5 mm square). If the center of the wipe mount is not aligned with the center of the deposit, the sample path may miss a portion of the deposit, and the reproducibility and the collection would suffer. Another potential issue with the TL-slip/peel

tester, is the flexible wire that holds the wipe mount in place. If the wire is not properly straightened,⁴⁶ the wipe will not have a smooth motion as the surface is translated underneath it. This zig-zagging movement could also negatively impact the CE%. While useful, potential human error and the linear sample path of the TL-slip/peel tester restrict the parameters that can be tested with this instrument.

This work expands upon the current capabilities of the TL-slip/peel tester, to see if additional parameters could be reliably studied. A commercial-off-the-shelf (COTS) 3D printer (Robo3D) was repurposed to use the XYZ stage for controlled placement of the wipe mount and two-dimensional wipe-sampling. A direct comparison of the TL-slip/peel tester to the 3D printer was completed and the capabilities of the system were explored. Previously studied parameters, (test surfaces, forces of collection, and linear sample paths) as well as the additional two-dimensional wipe-sampling feature of the 3D printer were investigated.

Experimental methods

Materials

Wipes and test surfaces—Commercial wipes were used as received and included Nomex® wipes (Smiths Detection, Edgewood, MD) and PTFE coated fiberglass woven wipes (Teflon) (DSA Detection, North Andover, MA). Test surfaces investigated included ABS plastic with an industry standard textured hair-cell finish (TAP plastics, Seattle, WA), ballistics nylon fabric (Seattle Fabrics, Seattle, WA), cardboard (cut from a shipping box), packaging tape (Scotch shipping tape commercial grade 3M 3750), synthetic leather (fabric.com), and 304 stainless steel with brushed (number 4) surface (Stainless Supply, Monroe, NC) (Figure S1). All materials were cut into 15 cm by 15 cm or 25 cm by 15 cm pieces to fit the platforms of the wipe-sampling instruments.

Wipe mount system—A wipe mount system developed in-house was adapted to fit both instruments, and is shown in Figure S2. The wipe mount system was fabricated to mimic the wipe clamp of sampling wands currently used in screening environments that can accommodate the different wipes, which come in many shapes and sizes. While a large section of the wipe could be used, the goal was to mimic field collection. The typical trace explosive detection instruments have a relatively small target area of the wipe that is exposed to the detector. Therefore, the wipe is secured between two pieces of plastic with a circular 3 cm diameter section exposed to the test surface.

Wipe-sampling Instruments—To compare to the current instrumentation used for standardized wipe-sampling for trace explosives in an operational setting, a TL-slip/peel tester (Imass TL-2200, Accord, MA[‡]) was used without modification. A COTS fused deposition modelling (FDM) 3D printer, the Robo3D (San Diego, CA[‡]), was repurposed to use its XYZ stage and the print head was re-engineered to hold the wipe mount. Custom control software (LabVIEW, Austin TX) was also written for the Robo3D to allow for the

[‡]Certain commercial equipment, instruments, or materials are identified in this document. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products identified are necessarily the best available for the purpose.

wipe mount to follow a well-defined linear, square, or serpentine sampling pattern. Movement of the platform and wipe mount was controlled with G-code commands⁵⁰ from a host computer to the microcontroller of the 3D printer. The applied load for both wipe-sampling instruments was governed by the weight of the wipe mount and was adjusted by adding or removing steel masses. A key difference between the two instruments was the components that were moving during the wipe-sampling experiment. For the TL-slip/peel tester, the wipe mount was held stationary by a flexible wire connecting the mount to the TL-slip/peel tester while the test surface moved in a linear direction with the translatable platen of the tester. The Robo3D instrument has two-directional wipe capabilities; the test surface moved along the x-axis while the wipe mount moved along the y-axis. Figure 1 depicts a schematic for a single collection event and images of the wipe mount and wipe-sampling instruments are shown in Figure S2: TL-slip/peel tester (A.) and Robo3D (B.).

Sample Preparation

Recent work by Verkouteren *et al.*^{45,46} developed protocols for preparing trace explosive samples for dry-transfer to a test surface and measuring collection by wipe-sampling from the surface. These protocols were followed for this work. Briefly, a solution of RDX was printed onto PTFE thin films using drop-on-demand (DOD) inkjet printing. Once the samples dried, deposits of RDX were left which, once transferred to a test surface (described below), simulated actual residues found in field wipe-sampling scenarios. A wipe-sampling instrument was used to collect the RDX deposits onto a wipe. The RDX was then extracted from both the starting thin film and the wipe and analyzed by electrospray ionization-mass spectrometry (ESI-MS) to determine the transfer and collection efficiencies, respectively.

Drop-on-demand (DOD) Inkjet Printing—To create sampling materials, DOD inkjet printing was used to deposit approximately 350 ng of RDX onto PTFE thin films (Bytac) with an aluminum foil backing (Bytac Bench and Shelf Protector Sheets; SPI Supplies and Structure Probe, West Chester, PA) in an 8 × 8 array. The solution used for printing of the RDX was used as received (1000 µg/mL in acetonitrile, 1.2 mL/ampoule, Cerilliant Corp., Round Rock, TX).

Dry-transfer procedure—Ethyl alcohol, (ACS reagent > 99.5 %, Sigma-Aldrich, St. Louis, MO) was used to clean the test surfaces prior to each transfer. A template (see Figure 2) was placed on top of the test surface to reproducibly transfer particles to the same location. The Bytac containing 350 ng of RDX was placed on top of the surface within the template, analyte side down. A finger was placed behind the deposit and, using a minimum of 10 N of pressure, the Bytac was translated 2 cm to 3 cm along the sampling path to transfer the RDX to the test surface. Surface striations that run the length of the test surface aid in removal of the RDX particles from the Bytac during dry-transfer. Thus, to optimize dry-transfer to the steel test surface, the Bytac was translated perpendicular to the striations and therefore perpendicular to the sampling path keeping within the template.

Surface wipe-sampling

After dry-transfer, the test surface was adhered to the translatable platen of the TL-slip/peel tester or to the xy-platform of the robo3D wipe-sampler with double sided tape. A single

strip of tape was placed perpendicular to the sampling path and was used to secure the edge of the test surface where the sampling path began. This allowed for the fabrics (ballistics nylon and synthetic leather) to stretch along the wipe path and prevented them from bunching up during the wipe-sampling. The collection wipe was secured in the wipe mount system with a circular 3 cm diameter section exposed for collection. The wipe mount was positioned to initially meet the test surface and immediately pass through the area of dry-transfer (Figure 2). The selected steel weight was attached to the top of the mount and the mount or test surface was translated to collect the RDX deposit. All experiments were repeated with a minimum of six trials and used the following wipe-sampling conditions unless otherwise noted: dry-transfer of 350 ng deposits of RDX, a Nomex® wipe, a translational velocity of 5 cm s⁻¹, an applied load of 660g, and a linear travel distance of 12 cm.

Comparison of wipe-sampling instruments—ABS plastic and ballistics nylon surfaces were investigated using the TL-slip/peel tester or the Robo3D with an applied load of 660 g for ABS plastic or 1060 g for ballistics nylon.

Wipe-sampling different test surfaces—ABS plastic, ballistics nylon, cardboard, packaging tape (a single piece of packaging tape placed over a strip of cardboard), synthetic leather, and stainless steel (sampling path parallel to step heights) were investigated using the Robo3D.

Wipe-sampling using a wide range of applied loads—ABS plastic, synthetic leather, and ballistics nylon were investigated using the Robo3D and an applied load of 260 g, 460 g, 660 g, 860 g, or 1060 g.

Wipe-sampling over different distances—ABS plastic was investigated using the Robo3D and a linear travel distance of 5 cm, 12 cm, or 20 cm.

Wipe-sampling using different wipe parameters—ABS plastic was investigated using the Robo3D and a Nomex® or Teflon wipe, an applied load of 260 g, 460 g, 660 g, 860 g, or 1060 g with the following test surface modifications (Figure 5 schematic): artificial sebum/modified fingerprint mixture not added (None), 2 µL artificial sebum (2 µL AS) prepared using the protocol recently developed by Sisco *et al.*,⁵¹ 10 µL artificial sebum (10 µL AS), or 10 µL of a modified fingerprint mixture (10 µL MFM) comprised of 200 µL squalene, 200 µL palmitoleic acid, 50 mg pentadecanoic acid in 6 mL hexane.

Wipe-sampling in two dimensions- pattern—ABS plastic was investigated using the Robo3D and a total travel distance of 12 cm following a single 12 cm line, an L-shape (6.0 cm per side), a U-shape (4.0 cm per side), or a square (3.0 cm per side) (Figure 6 schematic). To remove any effect from the hair-cell texture of the ABS plastic, the patterns were sampled in two orientations (0° and 90°).

Wipe-sampling in two dimensions-back and forth—ABS plastic was investigated using the Robo3D and a total travel distance of 20 cm in a linear or a 47 cm × 30 cm serpentine pattern (Figure 7 schematic).

Wipe-sampling in two dimensions-lifting the wipe mount—ABS plastic was investigated using the Robo3D and a total travel distance of 20 cm following a single 20 cm line, three 6.7 cm lines, four 5.0 cm lines, or five 4.0 cm lines (Figure 7 schematic).

Extraction and analysis

Extraction of both the wipe and used Bytac were completed to calculate the CE% of the wipe under various conditions. Collection wipes (Nomex® or PTFE-coated fiberglass) were cut to reduce the size to only the area that was presented to the test surface. The trimmed wipe was then placed into a 2 mL glass vial and 1 mL of methanol (Chromasolv grade, Sigma-Aldrich, St. Louis, MO[‡]) containing approximately 25 ppb isotopic RDX (*i*-RDX) (Cambridge Isotope Laboratories, Andover, MA) was added to extract the particles from the wipe. The vial was then capped, vortexed at 3,000 rpm for 30 s, and set aside for analysis. The *i*-RDX internal standard, containing 99% ¹³C₃¹⁵N₃ substitutions on the ring of the RDX molecule, provided a mass 6 Da higher than the analyte of interest. The Bytac was extracted by flowing 1 mL of methanol containing the same *i*-RDX internal standard over the surface and into a 2 mL vial. Additionally, a set of unused samples printed onto Bytac were extracted to obtain the baseline amount of RDX present on the Bytac after DOD inkjet printing.

Extracts were analyzed using electrospray ionization-mass spectrometry (ESI-MS). A Thermo Ulti-Mate 3000 LC system (Thermo Scientific, Waltham, MA[‡]) was used to provide solvent flow and sample injection. A mobile phase of 100 % methanol at 0.5 mL min⁻¹ was used, with a sample injection volume of 5 µL. The LC system was coupled with a JEOL JMS-T100LP mass spectrometer (JEOL USA, Peabody, MA[‡]). The mass spectrometer was operated in negative mode, scanning *m/z* 250 to *m/z* 300 at 0.5 s scan⁻¹. Other mass spectrometer settings included a needle voltage of -2,000 V, an orifice 1 voltage of 20 V, a ring voltage of 5 V, an orifice 2 voltage of 5 V, a peaks voltage of 2,000 V, and nitrogen as the desolvating and nebulizing gas.

Quantification of the extracts was completed by integrating the peak areas of *m/z* 284 ([RDX+NO₃]⁻) and *m/z* 290 ([*i*-RDX+NO₃]⁻) and comparing the ratio (*m/z* 284 : *m/z* 290) of the peak areas to a 13-point calibration curve. The masses of RDX remaining on the Bytac and collected onto the corresponding wipe of a given wipe-sampling run were then used to calculate the transfer and collection efficiencies of the wipe, as shown in the following equations:

$$\text{Transfer Efficiency}(\%) = \frac{(RDX_{\text{Baseline}} - RDX_{\text{Bytac}})}{RDX_{\text{Baseline}}} * 100 \quad (\text{Eqn 1})$$

$$\text{Collection Efficiency}(\%) = \frac{RDX_{\text{Wipe}}}{(RDX_{\text{Baseline}} - RDX_{\text{Bytac}})} * 100 \quad (\text{Eqn 2})$$

Where $RDX_{Baseline}$ is the amount of RDX extracted from unused printed Bytac, RDX_{Bytac} is the amount remaining on the Bytac after dry-transfer, and RDX_{wipe} is the amount of RDX collected onto the wipe. Measured extraction efficiencies from both the Bytac and the Nomex were 98% or greater and therefore were not included as a parameter in the equation.

Results and Discussion

Comparison of wipe-sample instruments

To help address some of the issues of the current state of the art TL-slip/peel tester, a Robo3D (COTS 3D printer) was converted into an automated wipe-sampling instrument. To determine if this configuration produced results comparable to current technology, an initial comparison of the Robo3D and TL-slip/peel tester was completed using ABS plastic and ballistics nylon as test surfaces. Table 1 shows the collection efficiencies of RDX using Nomex® wipes obtained by the TL-slip/peel tester and Robo3D. The two wipe-sampling instruments provided nearly identical collection efficiencies for the two examined surfaces (1.4 % difference for ABS and 3.7 % difference for ballistics nylon). Though the absolute CE% of the two techniques was similar, the relative standard deviation (RSD) from ABS using the TL-slip/peel tester was 24 %, compared to roughly half that with the Robo3D (13 % RSD). This reduction in RSD indicates that the Robo3D wipe-sampling instrument can result in greater reproducibility in collection.

The manual placement of the wipe mount when using the TL-slip/peel tester introduces human error to the collection. The Robo3D system effectively minimizes the error with the software controlling the placement of the wipe mount, resulting in better alignment with the dry-transferred deposit. The software of the Robo3D also controls the movement of the wipe mount along a set of brackets (see Figure S2), which allows for a smooth translational movement. This control eliminates potential zig-zag movement which is present in the TL-slip/peel tester due to the flexible wire that holds the wipe mount in place.

The results of this evaluation indicate that not only was the Robo3D comparable to the TL-slip/peel tester in terms of CE%, but the Robo3D is an improved wipe-sampling instrument. The Robo3D allows for wipe-sampling collection across a 2-dimensional surface with repeatable force, distance, speed, and direction. With software controlling the placement and translational movement of the wipe mount, the Robo3D can provide a more reproducible measurement. From here, the wipe-sampling capabilities (test surface, applied load, and 2D wipe patterns) of the Robo3D were further investigated.

Wipe-sampling different test surfaces

Wipe-sampling is commonly encountered in many screening environments; therefore, the platform of the wipe-sampling instrument used for research must accommodate a wide variety of surfaces commonly encountered in the field. To evaluate whether the Robo3D could sample commonly screened surfaces, collection of RDX off multiple test surfaces was investigated. Six test surfaces were chosen, based on relevance to screening environments, and included: ABS plastic and ballistics nylon fabric (to simulate luggage), cardboard and packaging tape (to simulate cargo), synthetic leather (to simulate luggage and clothing), and

stainless steel (to simulate vehicles). Both transfer efficiency onto the test surface and CE% from the surface to the wipe were calculated (Figure 3).

Figure 3A shows the range of potential surfaces that can be examined and highlights the influence the test surface has on collection. The CE% of RDX using Nomex® wipes ranged from 10 % (ballistics nylon) to 44 % (ABS). The collection efficiencies obtained with these surfaces agree with wipe-sampling studies previously reported: 17 % for ballistics nylon and 37 % for ABS plastic.²⁴ Relatively smooth surfaces (packaging tape, synthetic leather, and steel) have comparable collection efficiencies (30 %) under the conditions investigated here. The CE% appears to decrease as texture along the sampling path is introduced. Though the steel has striations, the sampling path was parallel to the step heights and therefore the CE% from this surface was not affected by this texture. Cardboard, with its fibrous macro-texture, induced an approximate 10 % reduction in CE% compared to the smooth surfaces. Poor collection was achieved off ballistics nylon, likely due to the woven pattern made up of many fibers. Interestingly, the test surface with the most exaggerated texture (ABS plastic: 200 µm height profile)²⁴ had the most efficient collection. The height difference on the surface of ABS plastic appears to be drastic enough to limit the interaction with the wipe during sampling. After dry-transfer, the vast majority of the RDX particles are on the raised surface, easily accessible to the wipe upon initial contact.⁴⁵ Only half of the surface is estimated to be accessible to the wipe, and therefore the amount of intimate contact after the collection is drastically reduced, thus decreasing re-deposition. The texture as opposed to the flexibility or hardness, or even the chemical composition, of the test surface appears to have the most significant impact on the CE%. Future work will focus on better understanding the effect the chemical and physical properties of a surface have on the collection by wipe-sampling.

The Robo3D, like the TL-slip/peel tester, can accommodate a wide variety of surfaces, hard and soft, rigid and flexible, textured and smooth. As long as the surface can be cut to fit the platform and can be secured by double sided tape, either instrument can be used to determine the CE% of wipe-sampling. The TL-slip/peel tester platform is 46 cm long and can be used for larger distances for wipe-sampling but is limited to a single continuous linear collection before the user must adjust the wipe mount and test surface. The Robo3D platform (20 cm × 20 cm) has a maximum single linear collection distance of 20 cm; however, the xy-platform has the added 2D-wipe capabilities, which were investigated in a subsequent study. One limitation for both instruments is the lack of ability to investigate complex surfaces as the wipe mounts can only interrogate a flat surface. To better simulate field sampling, such as the inside of a suitcase, the wipe mount would need the ability to change orientation, an adaptation that has not yet been realized.

While the transfer efficiency (Figure 3B) spanned a wide range (24 % to 97 %), it is incorporated into the calculation for CE% and therefore is already accounted for. Packaging tape had a significantly lower transfer efficiency than other surfaces, which can be attributed to its smoothness. To overcome the poor transfer of RDX deposits to the tape, a 2 cm × 2 cm section of the tape was roughened by dragging sandpaper across the surface perpendicular to the sampling path where the dry-transfer takes place. By roughening this small area, the transfer efficiency to the tape was increased by a factor of 3.8, with the only effect on

collection being an increase in reproducibility (Figure 3A). Due to the high CE% of ABS plastic, and the prevalence of synthetic leather and ballistics nylon in literature, these surfaces were used for subsequent investigations of the capabilities of the Robo3D wipe-sampling instrument.

Wipe-sampling using a wide range of applied loads

One parameter that is commonly examined in wipe-sampling research is the effect the applied force has on the collection of trace residues. The extent to which the force affects the collection is arguably not fully understood, as some previous work shows only a modest increase^{36,44} in CE% with higher applied loads, while another shows a clear linear increase.²² These studies often focus on limited parameters, such as only two applied loads, or only one wipe-test surface interaction. To better understand the magnitude of effect this parameter has on collection, a wipe-sampling instrument must be able to apply a wide range of forces. Like the TL-slip/peel tester, the applied load of the Robo3D is governed by the weight of the wipe mount and can be adjusted by adding or removing steel weights (Figure S2). To test these capabilities, an extensive study was completed for three test surfaces (ABS plastic, synthetic leather, and ballistics nylon) using five applied loads by the addition of up to four 200 g steel weights to the wipe mount system. This provided applied loads from 260 g (wipe mount with no additional weights) to 1060 g (wipe mount plus four steel weights). For reference, a mass of 1000 g to 2300 g represents an estimated 'firm' pressure.²¹

Figure 4 shows that an increase in the applied load increases the CE% of RDX over the load range studied; though, the magnitude is dependent on the test surface. For ABS plastic, a significant enhancement in collection is initially observed, increasing from 23 % at 260 g to 41 % at 460 g. Applied loads greater than 460 g provided little improvement, indicating a modest force will result in optimal collection. A similar sharp increase is observed for synthetic leather, but a maximum CE% of approximately 30 % was observed at the second highest applied load. While increasing the force of collection can enhance the CE% by up to a factor of 1.8, this increase is reached at a modest force, around 660 g, for the ABS plastic and synthetic leather. The applied load appears to play a more significant role in the collection of RDX from ballistics nylon. The CE% increased by a factor of 1.75, but not until the highest applied load was used. These results indicate that the force of the wipe-sampling does have a positive impact on the collection, though it is not consistent across different test surfaces. The applied load necessary to reach the upper limit of the CE% will depend on the wipe, the test surface, and the extent to which increasing the force increases the intimate contact between the wipe and the test surface. Initial intimate contact is needed for the analyte to be picked up and embedded in the wipe, while intimate contact after the collection could be detrimental due to re-deposition of the analyte back onto the test surface. The Robo3D wipe-sampling instrument has the capability to investigate a wide range of applied loads and is therefore comparable to the TL-slip/peel tester in this regard. For the remaining experiments, the 660 g applied load was used to compare to recent literature.^{45,46}

Wipe-sampling over different distances

Wipe distance is another important aspect of trace particle collection. In field wipe-sampling for trace explosives detection the location of the analyte is unknown. Therefore, the contact

between the wipe and test surface needs to be sufficient to increase the likelihood of the wipe encountering the analyte. However, extended contact can promote re-deposition of the analyte back onto the test surface, which will negatively affect the collection.^{36,43} Understanding this balance is key to improving CE% and determining whether multiple short wipes should be used to fully interrogate a test surface, or if one longer wipe is sufficient. To demonstrate the wipe capabilities of the Robo3D, three linear distances (5 cm, 12 cm, and 20 cm) were tested. A longer wipe distance, after contact with the analyte, did in fact negatively impact the collection. The CE% dropped from 50.8 % $\pm$ 5.9 (5 cm) to 37.8 % $\pm$ 6.8 (20 cm). Though this is a significant decrease, the ABS plastic has such a high relative CE% that the wipe distance may not be as important of a factor in wipe-sampling. The magnitude of the negative impact for test surfaces that exhibit lower collection will likely be greater. The 20 cm $\times$ 20 cm xy-platform of the Robo3D limits the ability to investigate this parameter at greater wipe distances without altering the wipe pattern. Work is currently being completed on a next generation wipe-sampling instrument to allow for longer linear and patterned wipe distances with the same highly controlled conditions as the Robo3D.

Wipe-sampling using different wipe parameters and different wipes

Real world screening consists of sampling from surfaces that are not in pristine condition and are contaminated with a wide range of environmental residues. Sebaceous materials left behind by a finger or hand print are common contaminants that have been briefly investigated for their effect on collection by wipe-sampling. Studies have shown that when particles are transferred onto test surfaces through simulated fingerprints the collection is affected.^{36,48,52} Different wipe-surface combinations have led to both increases and decreases in CE%; therefore, it is important to better understand this effect. A major benefit of using the Robo3D is the ability to wipe multiple lines without manually adjusting either the test surface or the wipe mount. With this added feature, the wipe-sampling path was altered (Figure 5) so that the wipe would travel over the test surface twice in a single collection event. The first pass would take the wipe through an area of simulated fingerprint residue deposited onto the test surface, to pre-contaminate the wipe with sebum, while the second pass would travel across the analyte deposited from dry-transfer to test the effect of a contaminated wipe. This pattern was chosen to eliminate any possibilities for direct contact between the artificial sebum and deposited analyte before either was collected onto the wipe. Initial studies (Figure S3) used artificial sebum (AS),⁵¹ however the presence of the additional chemicals caused significant competitive ionization in the ESI-MS signal, making quantification difficult. To combat this interference, but still understand the role of a wipe contaminated with chemicals left behind during fingerprint deposition, 10 μ L of a modified fingerprint mixture (MFM, 200 μ L squalene, 200 μ L palmitoleic acid, 50 mg pentadecanoic acid in 6 mL hexane) was used. The results, shown in Figure 5, indicate that the CE% of wipe-sampling can be enhanced by a factor of 1.5 with the presence of fatty acids, waxes, and oils that are likely to be encountered in the field. With the ability to wipe multiple lines, the Robo3D allows for the investigation of different test surface modifications. This will lead to a better understanding of how contamination on a test surface will affect the CE% and will help identify potential detection issues due to the increased chemical background in real world wipe-sampling.

Many different materials have been used as the wipe for this type of sampling, with cotton being the most widely used and Teflon, Nomex®, and paper based wipes becoming more popular.⁵³ Sampling wands currently used in screening environments can accommodate the different wipe materials. The wipe mount (Figure S2) of the Robo3D wipe-sampling instrument was fabricated to mimic the wipe clamp of sampling wands and therefore the ability to accommodate different wipe materials was investigated with and without the addition of 10 µL of MFM with an applied load of 460 g or 860 g. The CE% of the Teflon wipe with ABS plastic was also enhanced by a factor of 1.4 (Figure S4) when the wipe was contaminated by the MFM before traveling through the dry-transfer area, regardless of the applied load. The wipe mount of the Robo3D will accommodate multiple wipe types and therefore can be used to quickly screen new wipes developed to optimize collection under different wipe-sampling environments.

Wipe-sampling in two dimensions

Wipe-sampling in screening environments often utilizes a two-dimensional sampling path, though the effect of direction has not been fully understood. Previous directional studies have been completed using a sampling wand which is controlled by the user and can therefore wipe in any direction and pattern; however, there is little to no control over the applied load, the speed of the translational movement, or the exact distance travelled.⁴³ To isolate the effect of a single parameter such as the sample path, the other parameters need to be held constant throughout the entirety of the wipe sampling motion. The xy-platform of the Robo3D allows for wipe-sampling in two dimensions while keeping the other parameters constant. This allows the isolation of the sample path and the ability to investigate how this alone affects collection. Four patterns, each with a total wipe distance of 12 cm, were investigated (Figure 6): a straight line (12.0 cm), an “L” shape (6.0 cm per side), a “U” shape (4.0 cm per side), and a square (3.0 cm per side). To remove any effect from the hair-cell texture of the ABS plastic, all four patterns were sampled with the plastic orientated at either 0° or 90°. The average CE% of the eight wipe experiments (four patterns, two hair-cell orientations each) is 45.0 % ± 6.5 %. Figure 6 shows that not only does the orientation of the hair-cell texture of the ABS plastic have little effect, the wipe-sampling pattern also has little effect on the collection. Surfaces with directional topography, such as the stainless steel, can now be quickly interrogated to determine if the orientation should be considered in field screening.

In addition to the square, the Robo3D can wipe in a serpentine pattern. To test if this back-and-forth motion would negatively affect the collection, two wipe sets (linear and serpentine) were completed with a total travel distance of 20 cm. The serpentine pattern, (Figure 7) was configured so that the wipe would not pass over the same area twice. The collection efficiencies for the two sampling paths was very similar with a 3.4 % difference, below the standard deviations of the means (6.8% for linear and 4.5 % for serpentine). The back-and-forth motion of the serpentine pattern, a field relevant sample path, imparts no effect on the CE% under these conditions.

Another added feature of the Robo3D system is the ability to wipe multiple lines without the user having to reposition the test surface or the wipe mount. With this capability, it was

possible to test the effect of lifting the wipe and repositioning it over the test surface multiple times during a single wipe-sample. To investigate this parameter, lines of different lengths (4.0 cm to 20 cm) were sampled. Figure 7 shows that it isn't until the wipe path was broken into five separate lines of 4.0 cm each that the CE% starts to decline, though only slightly. The ability of the Robo3D to investigate an entire 20 cm × 20 cm test surface, with a wide variety of wipe patterns and orientations while keeping the wipe in contact with the surface or lifting it repeatedly allows for a thorough wipe-sampling investigation that truly represents field screening applications.

Conclusion

This work validates the use of a new instrument to better understand how different parameters affect the collection of trace explosives by wipe-sampling. A COTS 3D printer was repurposed for its XYZ stage which removed user error during the placement of the wipe mount and guaranteed a smooth translational movement while maintaining constant speed, force, distance, and direction. These factors improved the reproducibility of the CE% over the widely-used TL-slip/peel tester while maintaining less than 4 % difference by direct comparison. Like other wipe-sampling instruments, the repurposed 3D printer could accommodate a wide variety of wipe materials, test surfaces, and applied loads. The added features of the xy-stage allowed for a much greater variety of wipe sampling patterns (linear, serpentine, and square) as well as allowing for wiping multiple lines without the need for adjustments by the user. This new instrument allows for rapid wipe-sampling experiments to better understand the effect of different parameters, and therefore better prepare screeners in real-world wipe-sampling scenarios.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

Schematic of the TL-slip peel tester for key times during a single collection event. For the COTS 3D printer, the wipe holder (4) would move while the platform (5) remains stationary. Additionally, no restraining line (6) would be present.

Figure 2.

Template (left) for dry-transfer of RDX to test surface and a schematic (right) for wipe-sampling: (A) Sample path, (B) footprint of wipe start location, (C) dry-transfer location of analyte particles, (D) footprint of wipe end location, (E) test surface.

Figure 3. Collection efficiencies (A.) of RDX using Nomex® wipes and dry-transfer efficiencies (B.) of RDX with different test surfaces using the Robo3D system. Uncertainties represent one standard deviation of the mean.

Figure 4. Comparison of collection efficiencies of RDX using Nomex® wipes across a range of applied loads for ABS plastic, synthetic leather, and ballistics nylon test surfaces. A 100 g load is approximately equal to 1 N of force. Uncertainties represents one standard deviation of the mean.

Figure 5.

Comparison (left) of collection efficiencies of RDX using Nomex® wipes and the Robo3D wipe-sampler for different test surface conditions: artificial sebum/modified fingerprint mixture not added (None), addition of 10 µL of artificial sebum (10 µL AS), and addition of 10 µL of modified fingerprint mixture (10 µL MFM). Uncertainties represents one standard deviation of the mean. Schematic (right) for wipe-sampling: (A1) first pass of the sample path, (B1) footprint of wipe start location for the first pass, (E) location of the added artificial sebum or modified fingerprint mixture, (D1) footprint of wipe end location for first pass, (A2) second pass of the sample path, (B2) footprint of wipe start location for the second pass, (C) dry-transfer location of analyte particles, (D2) footprint of wipe end location for second pass.

Figure 6.

Schematic (top) for wipe-sampling (A) sample path, (B) footprint of wipe start location, (C) dry-transfer location of analyte particles, (D) footprint of wipe end location. Wipe patterns include (I) single 12 cm line, (II) L-shape with 6.0 cm per side, (III) U-shape with 4.0 cm per side, and (IV) square with 3.0 cm per side. Graph (bottom) of collection efficiencies of RDX using Nomex® wipes for these sample paths and orientations of the hair-cell pattern of ABS plastic using a Robo3D wipe-sampler. Uncertainties represent one standard deviation of the mean.

Figure 7. Schematic (top) for wipe-sampling (A) sample path, (B) footprint of wipe start location, (C) dry-transfer location of analyte particles, (D) footprint of wipe end location. Sampling paths include (I) single 20 cm line, (II) 4.7 cm $\times$ 3.0 cm serpentine, (III) 3 lines, 6.7 cm each, (IV) 4 lines, 5.0 cm each, and (V) 5 lines, 4.0 cm each. Graph (bottom) of collection efficiencies of RDX using Nomex® wipes for these sampling paths across ABS plastic using a Robo3D wipe-sampler. Uncertainties represent one standard deviation of the mean.

Table 1

Comparison of collection efficiencies of RDX using Nomex® wipes for wipe-sampling using either the TL-Slip/peel tester or the Robo3D. Uncertainties represent one standard deviation of the mean.

Test Surface	Applied Load	Collection Efficiency (%)	
		TL-Slip/peel	Robo3D
ABS Plastic	660 g	44.3 ± 10.5	43.7 ± 5.5
Ballistics Nylon	1060 g	13.3 ± 2.3	13.8 ± 1.8