

**TECHNICAL NOTE****QUESTIONED DOCUMENTS**

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## Analyzing Brazilian Vehicle Documents for Authenticity by Easy Ambient Sonic-Spray Ionization Mass Spectrometry\*

**ABSTRACT:** Using desorption/ionization techniques such as easy ambient sonic-spray ionization mass spectrometry (EASI-MS), it is possible to analyze documents of Brazilian vehicles for authenticity, providing a chemical profile directly from the surface of each document. A method for the detection of counterfeit documents is described, and the falsification procedure is elucidated. Forty authentic and counterfeit documents were analyzed by both positive and negative ion modes, EASI(±)-MS. EASI(+)-MS results identified the presence of (bis(2-ethylhexyl)phthalate plasticizer and of dihexadecyldimethylammonium biocide in both types of documents. For EASI(-)-MS results, the 4-octyloxybenzoic acid additive ( $[M + H]^+$ :  $m/z$  249) is present only in counterfeit documents. It was also found that counterfeit vehicle documents are produced via Laserjet printers. Desorption/ionization techniques, such as EASI-MS, offer therefore, an intelligent way to characterize the counterfeiting method.

**KEYWORDS:** forensic science, questioned documents, desorption/ionization techniques, ambient mass spectrometry, EASI-MS, Brazilian vehicle documents

The analysis of documents for authenticity is an important field in forensic sciences. In Brazil, a large number of counterfeiting methods have been developed and are applied for a variety of documents such as identity cards, tickets, passports, banknotes, driver licenses, and vehicle registration documents (1).

Simple sensorial tests are used by nonexpert people to detect counterfeit banknotes. Nevertheless, more sophisticated counterfeit notes often escape these subjective tests. An increasing number of security items such as sophisticated security papers, latent images, watermarks, magnetic strips, special printing techniques, holographs, and areas with infrared or UV light responses, which are systematically increasing production costs, are therefore being applied. Counterfeiting uses mainly computational reproduction methods, which include image-capturing in electronic media (scanners), processing (software) and printing (laser, ink-jet, off-set), or direct photocopy. Owing to the diversity of counterfeiting methods and their increasing dissemination and sophistication, and counter-reactions from the counterfeiters based on the knowledge of the security items employed, new security items and techniques must constantly be created or improved for the law enforcement agencies

to stay “one step ahead.” Although sensorial inspection of security items and optical evaluation of image quality and patterns can still detect most counterfeit banknotes, chemical analysis of banknotes, especially whether security items are elaborated based on chemical fingerprinting screening, may provide an automated, fast, and reliable (unbiased) approach, able to detect forgery of increasing quality with nearly unquestionable results (1). Chemical fingerprinting of banknotes could fulfill these requirements but it has been, however, only sporadically tested or applied by forensic laboratories.

Investigations into documents have been performed by analyzing the chemical composition of inks. Many analytical methods have been used, including infrared spectroscopy (2,3), Raman spectroscopy (4,5) thin-layer chromatography (TLC) (6), high-performance liquid chromatography (HPLC) (7,8), capillary zone electrophoresis (9,10), gas chromatography coupled to mass spectrometry (GC-MS) (11,12), electrospray ionization mass spectrometry (ESI-MS) (13), field desorption mass spectrometry (FD-MS) (14), matrix-assisted laser desorption ionization (MALDI-MS) (15,16), and laser desorption ionization mass spectrometry (LDI-MS) (16). In general, methods such as GC-MS, MALDI-MS, ESI-MS, FD-MS, HPLC, and TLC require sample preparation procedures (extraction of pigments or dyes) and, consequently, cause the destruction of material evidence.

Recently, a new class of ionization techniques for ambient mass spectrometry (17–28) has been developed. These techniques allow desorption, ionization, and mass spectrometry characterization of analytes directly from their natural surfaces and matrixes (29), in an open atmosphere and with little or no prior sample work up, becoming an attractive tool for direct characterization of questioned documents without their destruction. Among these techniques, easy

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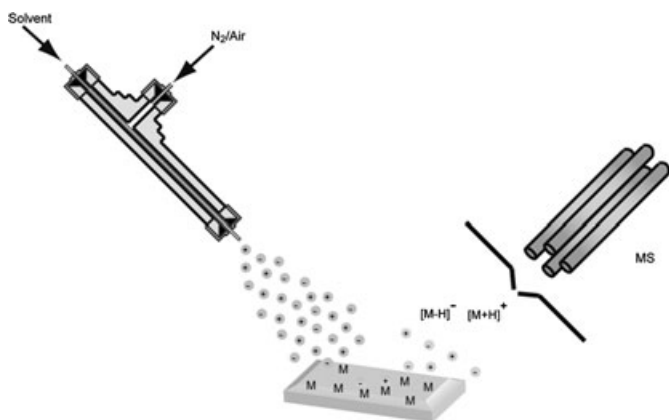


Fig. 1—Schematic of the EASI-MS system in operation on a surface solid. Sonic spray produces a bipolar stream of very minute charged droplets that bombard the solid surface causing desorption and ionization of the analyte molecules that rest on the target spot. Analytes are ionized often as either  $[M + H]^+$  or  $[M - H]^-$ , or both. EASI is assisted only by compressed nitrogen or air and causes no oxidation, electrical discharge, or heating interferences.

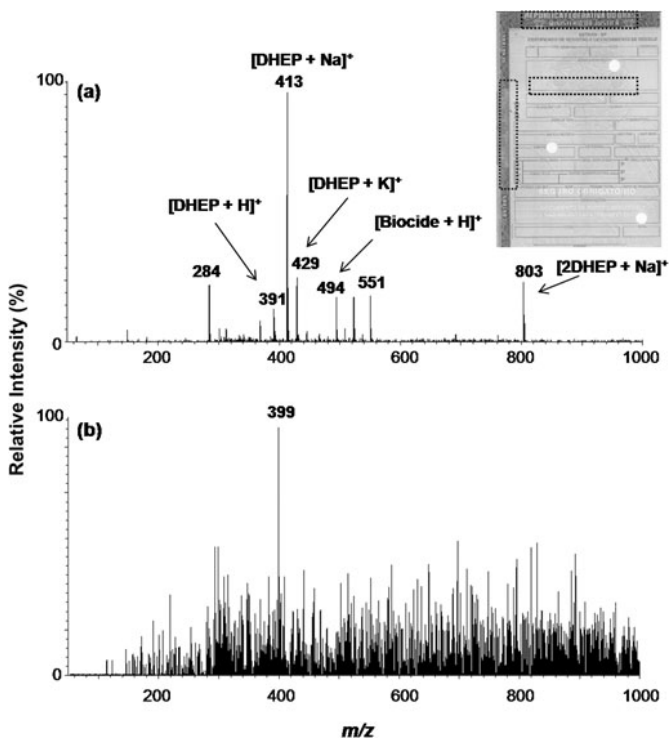


Fig. 2—(a) EASI(+)-MS and (b) EASI(-)-MS of authentic documents used for Brazilian vehicle registrations.

ambient sonic-spray ionization (EASI) is one of the simplest, gentlest, and most easily implemented techniques (30). An EASI source can be constructed, installed in few minutes (Fig. 1), and also operated with self-pumping provided by the Venturi effect (31). EASI relies on the forces of a high velocity stream of  $N_2$  (or even air) to accomplish analyte desorption and ionization by sonic-spray ionization (32). EASI has already been successfully tested with different analytes in different matrices and in various forensic applications, such as aging of ink writings on paper surfaces (33), perfumes (34), biodiesel (35), drugs of abuse (36,37), and identifying fake banknotes (1). EASI has also been coupled to TLC (38,39) and high-performance thin layer chromatography (40). EASI selectivity has been improved using molecularly imprinted polymers as selective surfaces (41). In this work, we investigated the applying of easy ambient sonic-spray ionization mass spectrometry (EASI-MS) for a rapid *in situ* analysis of questionable documents for official Brazilian vehicle registrations.

## Methods and Materials

HPLC-grade methanol, formic acid, and ammonium hydroxide were purchased from Burdick & Jackson (Muskegon, MI). Forty authentic and questionable documents were provided by the Criminalistic Institute of São Paulo State.

### EASI-MS

Experiments were performed on a single quadrupole mass spectrometer (LCMS-2010EV; Shimadzu Corp., Kyoto, Japan) equipped with a home-made EASI source. Acidified methanol (with 0.1% in volume of formic acid) at a flow rate of 20  $\mu\text{L}/\text{min}$  and compressed  $N_2$  at a pressure of 100 psi were used to form the sonic-spray used in the positive ion mode, EASI(+)-MS. For the negative ion mode, EASI(-)-MS, formic acid was substituted by ammonium hydroxide. The entrance angle of the capillary relative to the sample surface was *c.* 45°. Each document was directly analyzed by EASI-MS, without any sample treatment.

To confirm the structure of compounds found in the documents, the EASI source was coupled to ultrahigh-resolution and ultrahigh-accuracy Fourier transform-ion cyclotron resonance mass spectrometer (EASI-FT-ICR-MS; ThermoScientific, Bremen, Germany). Mass spectra were accumulated over 100 microscans, centered, and aligned using the XCALIBUR 2.0 software (ThermoScientific). The elemental composition of the compounds was attributed by the measurement of  $m/z$  values and double-bond equivalent (DBE) values.

## Results

Initially, the chemical profiles of authentic documents were obtained by EASI-MS in both ion modes (Fig. 2a,b). Three different positions on surface of the documents were analyzed and are illustrated in insert of Fig. 2a. In all cases, similar chemical profiles were obtained. The EASI(+)-MS fingerprint is shown in Fig. 2a,

Table 1—Exact mass of structures presents in Brazilian vehicle registration documents analyzed by EASI-FTMS.

Mode Ion	Precursor Ion $m/z$	Fragments (MS/MS) $m/z$	Structural Formula	DBE	Error (ppm)
Negative	249.1499	205 and 137	$[\text{C}_{15}\text{H}_{21}\text{O}_3 - \text{H}]^-$	5.0	1.21
Positive	391.2845	279, 167, and 149	$[\text{C}_{24}\text{H}_{38}\text{O}_4 + \text{H}]^+$	6.0	0.65
Positive	413.2665	—	$[\text{C}_{24}\text{H}_{38}\text{O}_4 + \text{Na}]^+$	6.0	0.75
Positive	429.2405	—	$[\text{C}_{24}\text{H}_{38}\text{O}_4 + \text{K}]^+$	6.0	0.75
Positive	494.5664	426, 408, 298, and 270	$[\text{C}_{34}\text{H}_{71}\text{N} + \text{H}]^+$	0	1.01
Positive	803.5435	735	$[\text{C}_{48}\text{H}_{76}\text{O}_8 + \text{Na}]^+$	12	0.37

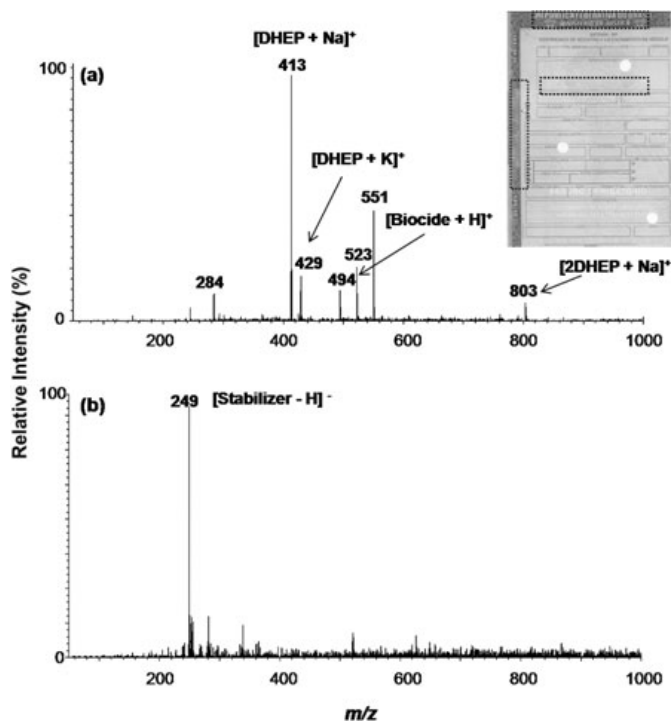


Fig. 3—(a) EASI(+)-MS and (b) EASI(-)-MS of counterfeit documents used in Brazilian vehicle registration.

where a series of ions of  $m/z$  284, 391, 413, 429, 494, 523, 551, and 803 was detected.

Acquiring mass spectra using the high-resolution and high-accuracy ICR cell of the linear ion trap Fourier transform mass spectrometry (FTMS) instrument, as well as examining the fragmentation profile as revealed by EASI-FTMS/MS (Table 1) allowed the identification of the diagnostic ions encountered in these samples.

Table 1 shows  $m/z$  values of diagnostic ions, their fragments, structural formula, DBE, and error predicted supplied by EASI-FTMS.

The EASI(+)-FTMS/MS results for the ion of  $m/z$  391 are in agreement with the bis(2-ethylhexyl)phthalate plasticizer, DHEP (Table 1). The fragments ions of  $m/z$  279 and 167 are formed via one and two neutral losses of the ethylhexyl group (112 Da), respectively. Subsequently, the ion of  $m/z$  167 losses  $H_2O$  to produce the ion of  $m/z$  149 corresponding to the acylium ion  $[HOO-CC_6H_4CO_2]^+$ . The ions of  $m/z$  413 and 429 correspond to sodium  $[M + Na]^+$  and potassium  $[M + K]^+$  adducts of the DHEP plasticizer, respectively. All species display DBE = 6. The ion of  $m/z$  803 is detected as a cluster of DHEP,  $[2DHEP + Na]^+$ , showing DBE = 12 (Table 1).

The EASI(+)-FTMS/MS results for the ion of  $m/z$  494 is in agreement with dihexadecyldimethylammonium biocide,  $[CH_3(CH_2)_{15}N(CH_3)_2(CH_2)_{15}CH_3]^+$ . The fragment ion of  $m/z$  270, a more abundant species, is formed via losing of  $CH_3(CH_2)_{14}CH_3$ . Quaternary ammonium compounds, such as the biocide dihexadecyldimethylammonium, are important classes of surfactants, being utilized in a wide variety of applications, such as fungicides, bleaching activators, biocides, algacides, softeners, and conditioners (1, 42).

The ions of  $m/z$  284, 523, and 551 also appear in the background of EASI(+)-MS when only methanol is used. Therefore, these ions are common laboratory contaminants from detergents (1). The EASI(-)-MS fingerprinting for the authentic documents is shown in Fig. 2b. No compound is detected in the negative ion mode.

The EASI( $\pm$ ) MS fingerprints for counterfeit documents are shown in Fig. 3a,b. The chemical profile for authentic and counterfeit documents (Figs 2a and 3a) are similar when comparing the EASI(+)-MS results. A specific and distinct chemical profile, however, is obtained with EASI(-)-MS when analyzing the counterfeit samples. An intense and unique ion of  $m/z$  249 was observed only for counterfeit documents (Fig. 3b).

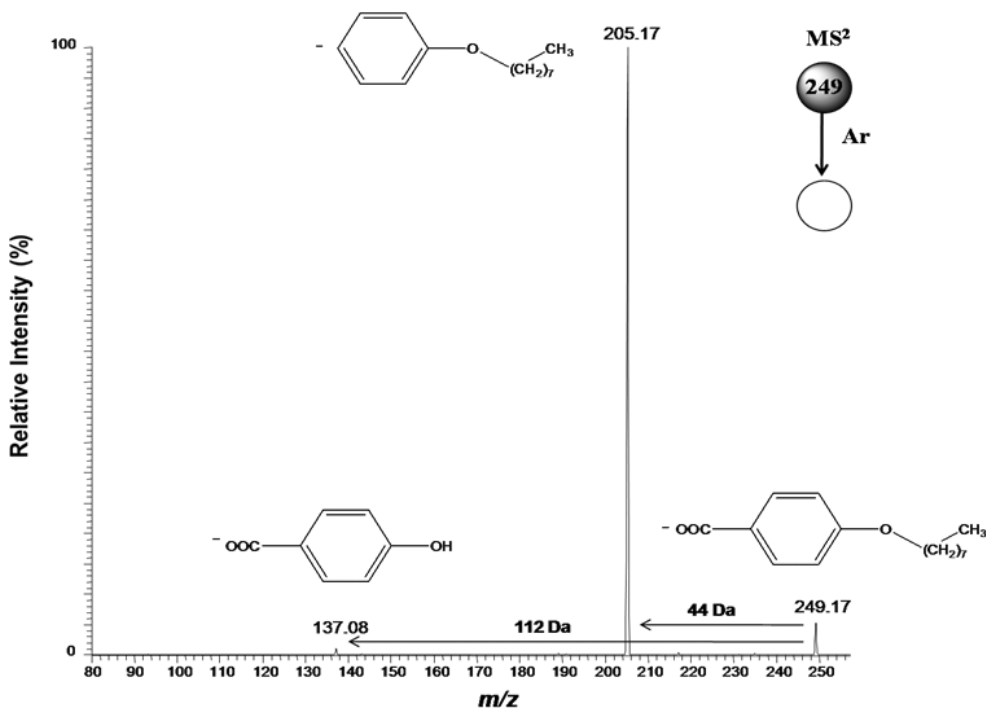


Fig. 4—EASI(+)-MS/MS of  $m/z$  249.

The EASI(-)-MS/MS results of ion of  $m/z$  249 are shown in Fig. 4. Two fragment ions were observed, an intense ion of  $m/z$  205 and another of  $m/z$  137. The ion of  $m/z$  205 is formed via loss of  $\text{CO}_2$  (44 Da). The ion of  $m/z$  137 is formed via loss of the aliphatic part ( $\text{CH}_2=\text{CH}(\text{CH}_2)_5\text{CH}_3$ ) (112 Da). Both are produced from the precursor ion,  $m/z$  249. Complementary information is also obtained when analyzing the structural formula and DBE predicted by EASI-FTMS, being  $[\text{C}_{15}\text{H}_{21}\text{O}_3]^-$  and 5.0, respectively. These results indicate the presence of the compound 4-octyloxybenzoic acid. Generally, printer ink composition is comprised of a colorant, an optional viscosity modifier, an optional conductivity enhancing agent, and an optional second acid. Among these substances, the 4-octyloxybenzoic acid acts as an optional second acid, being a stabilizer added in low concentration, no more than 1 wt%. As a consequence, it can be inferred that the documentation centers that produce authentic Brazilian documents do not use derivatives of benzoic acid in the production of these documents. The presence of this ion in counterfeit samples indicates the counterfeiting method.

To identify the possible origin of the ion of  $m/z$  249, ink compositions used in different printers were analyzed. Figure 5a-c shows EASI(-)-MS fingerprints for three types of laboratory-made

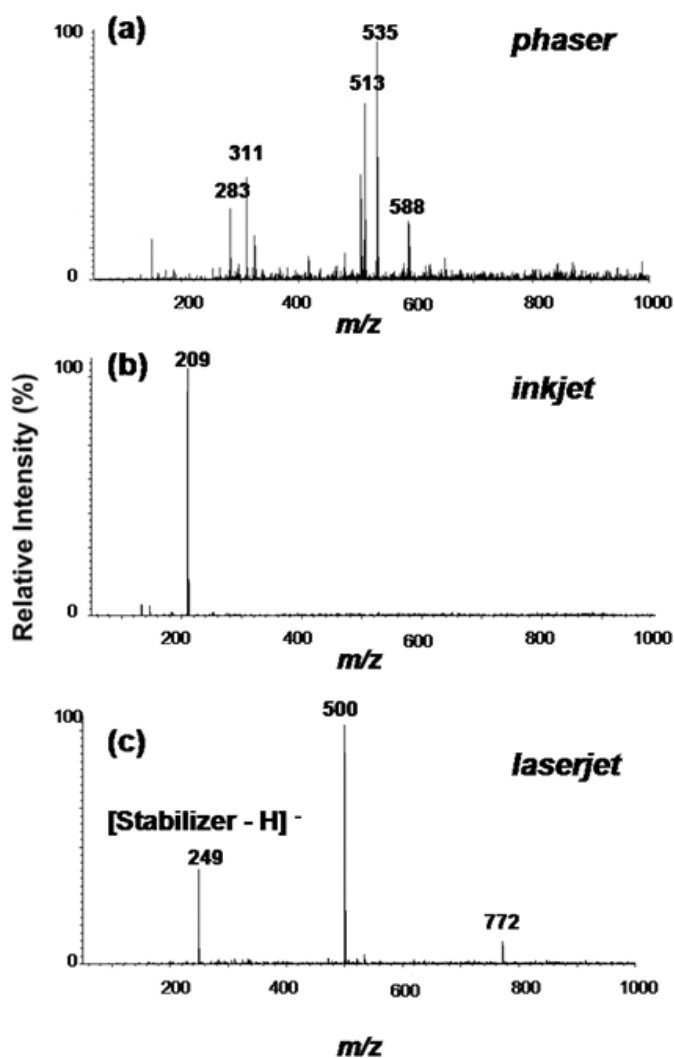


Fig. 5—EASI(-)-MS fingerprints of laboratory-made documents similar to those used for Brazilian vehicle registrations, prepared using Phaser (a); Deskjet (b); and Laserjet (c) printers.

counterfeit samples: Phaser (5a), Deskjet (5b), and Laserjet (5c). The EASI(-)-MS for the Phaser printer ink shows intense ions of  $m/z$  283, 311, 513, 535, and 588. Analyzing a Deskjet printer ink, a unique intense ion of  $m/z$  209 is observed. Finally, EASI(-)-MS results for Laserjet printer ink show ions of  $m/z$  249, 500, and 772. The EASI(-)-MS results obtained for counterfeit samples studied herein indicate that they are falsified using Laserjet printers.

## Discussion

EASI-MS provides a direct, robust, and reliable chemical fingerprinting method for a fast and nondestructive screening of documents used for Brazilian vehicle registrations. Chemical profiles were obtained for authentic and counterfeit documents. EASI(+)-MS results detected the presence of bis(2-ethylhexyl)phthalate plasticizer in several forms:  $([\text{M} + \text{H}]^+)$ :  $m/z$  391;  $([\text{M} + \text{Na}]^+)$ :  $m/z$  413;  $([\text{M} + \text{K}]^+)$ :  $m/z$  429; and  $([2\text{M} + \text{Na}]^+)$ :  $m/z$  803. Also, the biocide dihexadecyldimethylammonium  $([\text{M} + \text{H}]^+)$ :  $m/z$  494) was detected. A faster and clearer distinction among the documents is reached when the chemical profiles of documents in the negative ion mode, EASI(-)-MS, are obtained. For counterfeit documents, the 4-octyloxybenzoic acid compound  $([\text{M} - \text{H}]^-)$ :  $m/z$  249) is detected, being used as a stabilizer in printing ink. It was also found in homemade documents produced from Laserjet printers. Desorption/ionization techniques offer, therefore, the origin of the diagnostic ions identifying the counterfeiting method.

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