Chemometric and trace element profiling methodologies for authenticating, crossmatching and constraining the provenance of illicit tobacco products

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ABSTRACT

Background

Illicit tobacco products impact negatively on public health. Counterfeits and cheap whites as well as legal brands smuggled from countries not adopting track and trace technologies will require novel forensic tools to aid the disruption of their supply chains.

Methods

Datasets of trace element concentrations in tobacco were obtained using X-ray fluorescence spectrometry on samples of legal and illicit products mainly from Europe. Authentic and counterfeit products were discriminated by identifying outliers from datasets of legal products using Mahalanobis distance and graphical profiling methods. Identical and closely similar counterfeits were picked out using Euclidean distance, and counterfeit provenance was addressed using chemometric methods to identify geographic affinities.

Results

Taking Marlboro as an exemplar the major brands are shown to be remarkably consistent in composition, in marked contrast to counterfeits bearing the same brand name. Analysis of 35 illicit products seized in the EU indicates that 18 are indistinguishable or closely similar to Marlboro legally sold in the EU while 17 are sufficiently different to be deemed counterfeit, among them being two counterfeits so closely similar that their tobaccos are likely to come from the same source. The tobacco in the large majority of counterfeits in this dataset appears to originate in Asia.

Conclusions

Multivariate and graphical analysis of trace elements in tobacco can effectively authenticate brands, crossmatch illicit products across jurisdictions and may identify their geographical sources.

WHAT THIS PAPER ADDS

- Illicit tobacco significantly undermines tobacco control measures aimed at encouraging smokers to quit and reducing under-age access to products.

- Track & trace is a key policy in the FCTC protocol to eliminate the illicit trade but these technologies will not be implemented in counterfeit and cheap white products, nor necessarily included in legal products smuggled from countries not ratifying the protocol.

- Major cigarette brands such as Marlboro are shown to have highly consistent trace element compositions that do not change significantly over periods of years but may differ between geographic regions.

- Chemometric and trace elements profiling of tobacco products lacking track and trace can be used to discriminate authentic from counterfeit products and can also crossmatch seizures of counterfeits from the same illegal source. Regional differences can help constrain the provenance of tobacco.

- These forensic tools can be applied independently of the tobacco manufacturers and could beneficially complement track & trace technologies in providing intelligence to disrupt the supply routes of illicit products.

BACKGROUND

Illicit tobacco

Tobacco quitting rates respond well to increasing price through taxation but this public health measure is undermined through recourse to illicit products by the many smokers who cannot afford to maintain their habit with legitimate brands.[1 2] At the same time unregulated sales of illicit tobacco make it easier for young people to start smoking. These issues were recognised in Article 15 of the WHO Convention on Tobacco Control (FCTC) that was elaborated in 2013 in the form of a protocol to eliminate illicit trade in tobacco products. [3 4] Much of the protocol focuses on the need for legitimate tobacco manufacturers to implement track and trace (T&T) technologies so that products evading taxation will be quickly and easily identifiable, at least until technologies are counterfeited. A shortcoming of this measure is that illicit products that do not deploy T&T will require other forensic and intelligence-gathering methods to disrupt their supply chains and distribution networks.[5-7]

In tackling the problem of illicit tobacco governmental agencies such as customs and border authorities, police and trading standards, typically rely on the manufacturers to establish the authenticity of seizures bearing their own brands, however there are circumstances when this leads to serious conflicts of interest for the tobacco industry. Examples include agreements that penalise tobacco companies when supply chains leak legal products into illicit distribution networks, including the European Union agreements with the four major tobacco manufacturers operating in the EU.[8 9]

Large discrepancies exist between the estimated share of illicit tobacco products published by KPMG on behalf of the tobacco industry (with industry involvement in authentication) and the official statistics of the UK Government.[10 11] This may be due in part to the use of very different methodologies but while the manufacturers are directly involved in generating the

raw data the conflict of interest will always lead to suspicion of industry statistics, whether justified or not.[12]

Tobacco in counterfeit cigarettes often contains higher levels of heavy metals than the products they mimic. [13-16] A wider range of trace elements may provide more sensitive and reliable authentication and provenance indicators than a small number of heavy metals. While requiring more time and laboratory resources than most T&T technologies chemical approaches offer the possibility of authentication judgments that are independent of packaging and avoid tobacco company involvement. Chemical profiling is also capable of contributing intelligence on the supply chains of illicit products and may also provide insights into cheap white production and distribution, although the focus of this paper is on illicitly manufactured products of major legal brands, i.e. counterfeits.

Origins of trace elements in tobacco

Tobacco hosts a wide range of inorganic elements (typically referred to as trace elements) that can amount to 5% or more of the total dry weight of tobacco and their presence is evidenced in the ash that remains after a cigarette is smoked.[17 18] Their origins are diverse. Soil and dust particles are the main sources of insoluble elements such as Si, Ti and oxidised Fe [19] whereas other elements tend to be more soluble and are varyingly taken up by the roots depending on the variety of tobacco and other factors. Among these are major elements such as K, Ca and P whereas some minor elements such as cadmium have a strong affinity for tobacco and preferentially accumulate in leaves and/or stems.[20] Some essential elements including K, P, S, Cl, and Ca may be further enriched by the application of fertilisers while others, notably the metals, may be enhanced in areas of industrial pollution.[18] Additives at the manufacturing stage might also contain trace elements included in mineral compounds

and salts (notably Ca, K, Sr and Rb).[21] The bulk trace element composition of tobacco products reflects these diverse origins.

Rationale for using trace elements in counterfeit identification

Most trace elements in tobacco derive primarily from environmental sources and agronomic practices leading to the hypothesis that different regions of the world will show distinctive patterns of trace element concentration reflecting local environments and agronomies. By contrast the major producers often export their cultivation practices to new countries and their blends normally involve several varieties that may be sourced globally. These factors largely obfuscate local signatures in the transnational brands of big tobacco companies and the blends are tightly controlled leading to highly consistent products. It is relevant here that tobacco used in counterfeits is rarely consistent. The evidence for these differences is presented below and supports the proposition that trace elements could provide a method for discriminating counterfeit from authentic products. This is tested by comparing a selection of trace elements in seizures of illicit products with a dataset of authentic products bearing the same brand name. If counterfeit cigarettes are manufactured from tobaccos of restricted geographical provenance then there is also a possibility that they may retain a trace element signature of their geographical source.

The large tobacco companies compete by offering distinctive products usually based on blends and flavourings.[22] When blends are distinctive and remain consistent over long periods of time their associated trace element compositions may be expected to behave similarly. This paper examines the potential of tobacco blends to yield reliable trace element signatures that can be used for authenticating, matching and provenancing cigarette products as an alternative to conventional authentication methods that are usually based on extrinsic features of packaging and stamps which may be more amenable to counterfeiting.

METHODS

Sampling and analysis

Four tobacco companies, namely Philip Morris International (PMI), Japan Tobacco International (JTI), British American Tobacco (BAT), and Imperial Tobacco Limited (ITL) are estimated to supply more than 80% of the legal cigarette market in the European Union. [23] Much of the following concerns the Marlboro cigarette brand (Philip Morris) that was selected for this study for the sole reason that it is the world's most popular brand in terms of sales. The brand is sold in various forms in most countries and is one of the most common targets for counterfeiting.[24] Large numbers of illicit cigarettes recovered from seizure operations in Europe and North America have been investigated in the laboratories at the University of St Andrews using packs supplied by several governmental agencies. A subset of 35 packs of Marlboro recovered in seizure operations and donated for study by two national agencies in the EU is used here to illustrate the authentication and provenance methodologies although their use does not imply formal accreditation by any of these agencies. It is not known when the seizures were made and many may predate the implementation of the Philip Morris-EU Agreement.[25]

Packs of legal brands were purchased at intervals over several years in large retail chains such as major supermarket stores and at airport shops in several EU countries. The observed consistency of profiles at brand level confirms that products acquired in this way are almost invariably authentic and obviated the need to acquire samples from the manufacturers. A separate collection was made of minor, non-transnational brands purchased in major tobacco-growing countries, the rationale being that such purchases are more likely to contain unblended tobacco of local origin.

Tobacco was extracted from the cigarette sticks in a pack, pulverised to fine powder and reconstituted as a pellet for polarised X-ray fluorescence analysis of 17 trace elements (Si, P, Cl, S, K, Mg, Ca, Ti, Mn, Fe, Cu, Zn, Br, Rb, Sr, Cd and Ba). Details of the method are given in the supplementary file on methodology along with accuracy and precision data for these elements and their limits of detection.

Graphical and statistical methods

Trace element profiles presented in this paper are generated by plotting the ratio of the concentration of each element in tobacco normalised to the same element in the reference material. The University of Kentucky's Research Cigarette 1R5F, created to represent US blends that yield low levels of nicotine, was arbitrarily selected as the reference tobacco.[26] The profile is constructed with trace elements along the X-axis in a sequence of increasing valence in the surface environment, first anions then cations. The Y-axis is the $\log_{10}$ value of the reference-normalised trace element concentration. A profile is generated for each tobacco by joining neighbouring data points with tie lines to produce a line graph (see examples in figures below). It is important to note that apparent variability in trace elements at lower concentrations appears somewhat greater on these log-ratio plots compared with the variability in higher concentration elements such as the essential elements K, Ca, P and S for similar differences in the absolute concentrations. More characteristics of these profiles are given in the supplementary file on methodology.

Multivariate statistical methods are widely used in the chemometric approach to investigating the structure of complex datasets, particularly in classifying objects. In this study counterfeit tobaccos are identified using Mahalanobis distance in order to estimate the probability of a pack belonging to a multivariate distribution of genuine packs using the distance between a pack and the distribution mean of genuine packs in multidimensional space. The Euclidean

distance coefficient is used to find the most similar pairs of cigarette packs in multidimensional space. The uniqueness of products from the major manufacturers was explored using discriminant function analysis (DFA) to test whether the products of different manufacturers are characterised by distinctive ranges of trace element concentrations.[27] Provenance was addressed using principal components analysis (PCA) to seek compositional patterns in tobacco that are associated with geographical regions, a widely-used chemometric technique in food and drink provenance.[28]

These multivariate data exploration techniques are based on latent variables created from the original trace element measurements. Whereas the uncertainties in these measurements are known (Supplementary Table 1) it is less straightforward to estimate uncertainties in the derived latent variables. A more complete description of the multivariate methods used in this study and how uncertainties are considered is provided in the Methodology supplementary file.

RESULTS

Distinctiveness of trace elements by manufacturer

An important assumption for trace element methods is that major brands are different but internally consistent products that will yield reliable trace element "fingerprints". It is useful first to consider whether the popular brands of the major manufacturers are distinct. Fig.1 shows the results of a discriminant function analysis for the major brands of the four main manufacturers sampled in the EU between 2006 and 2014. The plot shows each pack connected by tie line to the discriminant function means for each manufacturer. The first two discriminant functions for the sampled products of Philip Morris, Imperial Tobacco, British American Tobacco (Fig.1) shows that they have distinct trace element compositions while the products of Japan Tobacco overlap those of the other three manufacturers. Furthermore, the

Philip Morris brands Marlboro and L&M largely resolve into different areas (Fig.1). These findings support the concept that individual brands may have distinct trace element characteristics.

Consistency of individual brands

To test the degree of trace element consistency at brand level and to examine which elements contribute to any lack of consistency 33 profiles for the Marlboro brand (both red and gold pack variants) sampled from legal sources across the EU between 2004 and 2014 are compared in profile form (red lines in Fig.2). It appears that the profiles have remained broadly consistent over the decade with only Br, Cd and Ti showing a notable degree of variability (reasons are discussed below).

To facilitate comparison with illicit products a field is plotted that defines two standard deviations either side of the mean for each element over all 33 profiles and this is taken to represent 95% of the range of EU Marlboro sampled in that decade (yellow field in Fig.2). Comparing this field with eight profiles for Marlboro (both red and gold packs) purchased in the US during the same period it is clear that these products also have highly consistent profiles (blue lines in Fig.2) but are distinct from Marlboro marketed in the EU, the differences presumably reflecting the use of different blends and/or sources of tobacco. This evidence for consistency makes these trace element profiles suitable for use as fingerprints of brands and potentially of different production facilities for the same brand.

Use of profiles for authentication

Fig.3 shows the profiles for 35 seizures branded as Marlboro (red and gold packs) provided for study by two national customs authorities within the EU. 18 profiles (green lines) fit the field for authentic EU Marlboro defined in Fig.2 with more than two thirds of trace elements

falling inside the 95% boundary (yellow field) with the remainder being close. These are strong candidates for authentic products. The profiles of another 17 seizure packs (red lines) indicate poor fits with half or more trace element concentrations falling outside the 95% probability envelope. These are all strong candidates for counterfeit Marlboro. In support of this conclusion the tie lines that make up the red patterns also show reversals in slope compared with authentic products, for example Zn-Cd and Cd-Ba tie lines in Fig.2.

Mahalanobis distance estimates for each of the 18 seizure packs represented by the green line profiles on Fig.3 indicate that none is significantly different from the group of 33 authentic products shown on Fig.2 ($p<0.025$). In contrast the 17 seizure packs represented by the red line profiles of Fig.3 are all highly significant outliers from the authentic set with low probabilities that these differences arise by chance (Table 1). Assuming that the authentic samples are representative of EU Marlboro over the time interval during which the seizures were made then a combination of visual pattern comparison and statistical outlier recognition effectively discriminates authentic from counterfeit products.

Seizure ID	Apparent brand	Member State of seizure	Mahalanobis D (p<0.025)	% elements within ±2s of profile	Authenticity interpretation	Geographic affinity
S1	Marlboro (red)	A	Inlier	76	Very similar to authentic	EU Marlboro
S2	Marlboro (gold)	A	Inlier	88	Indistinguishable from authentic	EU Marlboro
S3	Marlboro (red)	B	Inlier	88	Indistinguishable from authentic	EU Marlboro
S4	Marlboro (gold)	B	Inlier	76	Very similar to authentic	EU Marlboro
S5	Marlboro (gold)	B	Inlier	100	Indistinguishable from authentic	EU Marlboro
S6	Marlboro (red)	B	Inlier	94	Indistinguishable from authentic	EU Marlboro
S7	Marlboro (gold)	B	Inlier	94	Indistinguishable from authentic	EU Marlboro
S8	Marlboro (red)	B	Inlier	71	Very similar to authentic	EU Marlboro
S9	Marlboro (gold)	B	Inlier	88	Indistinguishable from authentic	EU Marlboro
S10	Marlboro (red)	B	Inlier	82	Indistinguishable from authentic	EU Marlboro
S11	Marlboro (gold)	B	Inlier	71	Very similar to authentic	EU Marlboro
S12	Marlboro (gold)	B	Inlier	88	Indistinguishable from authentic	EU Marlboro
S13	Marlboro (gold)	B	Inlier	76	Very similar to authentic	EU Marlboro
S14	Marlboro (gold)	B	Inlier	88	Indistinguishable from authentic	EU Marlboro
S15	Marlboro (red)	B	Inlier	94	Indistinguishable from authentic	EU Marlboro
S16	Marlboro (red)	B	Inlier	100	Indistinguishable from authentic	EU Marlboro
S17	Marlboro (red)	B	Inlier	94	Indistinguishable from authentic	EU Marlboro
S18	Marlboro (gold)	B	Inlier	94	Indistinguishable from authentic	EU Marlboro
S19	Marlboro (red)	B	Outlier	47	Dissimilar to authentic, counterfeit	Uncertain
S20	Marlboro (gold)	A	Outlier	12	Dissimilar to authentic, counterfeit	Possibly Asia
S21	Marlboro (red)	A	Outlier	24	Dissimilar to authentic, counterfeit	Asia
S22	Marlboro (red)	A	Outlier	29	Dissimilar to authentic, counterfeit	Asia
S23	Marlboro (red)	A	Outlier	18	Dissimilar to authentic, counterfeit	Asia
S24	Marlboro (red)	B	Outlier	47	Dissimilar to authentic, counterfeit	Asia
S25	Marlboro (gold)	B	Outlier	35	Dissimilar to authentic, counterfeit	Asia
S26	Marlboro (gold)	B	Outlier	24	Dissimilar to authentic, counterfeit	Asia, same as 29

S31	Marlboro (red)	**B**	Outlier	24	Dissimilar to authentic, counterfeit	Asia
S32	Marlboro (red)	**A**	Outlier	29	Dissimilar to authentic, counterfeit	Asia
S33	Marlboro (red)	**A**	Outlier	18	Dissimilar to authentic, counterfeit	Possibly Asia
S34	Marlboro (gold)	**B**	Outlier	29	Dissimilar to authentic, counterfeit	None evident
S35	Marlboro (gold)	**B**	Outlier	24	Dissimilar to authentic, counterfeit	None evident

Table 1. Results for 35 seizures presented for study by two EU Member States (A & B). Red and gold refer to the dominant colour of a Marlboro pack. Inliers and outliers were identified using Mahalanobis distance. The %elements within ±2s of profile is based on a tally of the number of trace element data points falling within the yellow envelope of Figs 2 and 3. Geographical affinity for authentic products is based on profile comparison (e.g. Fig.2) and counterfeit affinity is based on Fig.4.

Matching illicit products

Applying the Euclidean distance coefficient to a large database of illicit packs reveals occasional clusters of two or more seizures, sometimes of different brands, that have essentially identical or closely similar profiles. This might be expected among products recovered in the same haul of illicit products but sometimes such matches are found in packs from different hauls and can then provide useful information on the distribution of the illicit product. Similarly seizures of shredded tobacco destined for use in illicit manufacture can sometimes be matched with finished products circulating in the illicit market.

The process of matching involves first seeking the closest statistical fits using the Euclidean distance coefficient and then visually comparing the patterns in pairs of profiles. If several possible matches are identified then they may be refined by rejecting statistical outliers using Mahalanobis distance [29].

Fig.3 shows a match of two Marlboro products seized in the EU. Using criteria described above for comparison with authentic EU Marlboro they are both clearly counterfeit. The profiles include error estimates based repeat analyses of ten separate packs of research cigarette 3R4F.[30] The visual match is very close although there is an obvious discrepancy in Cu (copper) and lesser discrepancies in Fe (iron), Sr (strontium) and Ti (titanium). Compared with the range of compositions in counterfeit products also shown in Fig.3 these discrepancies are relatively small and differences in Cu, Fe and Ti are probably be related to differential metal contamination by worn machinery, not uncommon in counterfeiting operations. The profile patterns match very well and these seizures are considered very likely to have been products of the same batch of tobacco.

Identifying geographical provenance

Principal components analysis was applied to trace element concentrations in local (non-transnational) cigarette tobaccos from a number of tobacco-producing countries enabling bivariate plots of principal component scores for each tobacco to be colour coded for regions where there is significant tobacco production and from where authentic products have been acquired. Fig.4 plots the first and third principal components that account for 49% of the variance in the original 17 variables. This projection is chosen as it maximises the resolution of Asian products, there being intelligence that some seizure products had probably originated in China.

Plotted on the same graph (Fig.4) are the fields for the authentic EU and US Marlboro packs used to create Fig.1. Also plotted are the 35 illicit Marlboro packs seized in the EU and analysed above for authenticity. 18 genuine Marlboro seizures (S1-18 in Table 1) fall within or at the margin of the field for authentic EU Marlboro whereas the 17 seizures considered counterfeit are widely distributed away from the authentic field. 12 seizures (S21-32) form a cluster along with S20 among a group of predominantly authentic non-transnational cigarette packs purchased in East Asia (mainly China). The only seizures that show no unique affinity are S19 that falls among brands from multiple regions and S34 & S35 that are not associated with any geographical region in the current dataset. These conclusions are collated along with authenticity interpretations in Table 1.

DISCUSSION

Consistency of profiles

Trace element profiles of Marlboro have remained remarkably consistent in the EU over at least the last 10 years demonstrating the value of intrinsic fingerprints, however a few trace

elements are more variable than others in the profiles shown in Fig.1, most notably Br (bromine), Ti (titanium) and Cd (cadmium). Variability in Br is particularly revealing. We monitored a steady decline in Br concentration for EU Marlboro products from ~100 to ~40 mgkg^{-1} between 2001 and 2010 since which time the concentration has remained relatively constant at ~35 mgkg^{-1}. The declining Br trend most likely reflects the Montreal Protocol to protect the ozone layer which required the phasing out of methyl bromide, a major fumigant used in tobacco cultivation but also an ozone-depleting compound.[31] Such influences underline the importance of monitoring tobacco products with time to take account of gradual or abrupt changes to blends or sources that may consequently affect their trace element profiles.

Variability in Ti is less clear but may be related to its common presence in dense metallic minerals (typically ilmenite and titanomagnetite) that may not distribute entirely homogenously during tobacco blending. The variability in Cd is related to its large relative analytical error when present in low concentrations (range of 0.5-6.1 mg/kg) approaching its XRF detection limit of 0.2 mg/kg (Supplementary Table 1). Despite these variabilities Br, Cd and Ti are retained in the fingerprint as they are still useful geographical discriminators, for example very low Br and very high Cd are currently strongly characteristic of many legal brands purchased in China.[32]

Validity of the method

A major obstacle to independent authentication of brands is the lack of access to proprietary information. The empirical approach adopted here is to use intrinsic properties of tobacco although it must be recognised that validation is difficult without involving the manufacturers. Between 2010-2014 large numbers of packs seized in the EU were submitted to this laboratory by OLAF, the European Commission's anti-fraud directorate (results are not

presented here for reasons of confidentiality). Authentication tests were carried out by the manufacturers and some of the same seizures were analysed independently by government customs laboratories while the University of St Andrews applied the methods described in this paper. The results were not shared but OLAF has reported that there were no discrepancies between the findings.[23]

Applicability to tobacco control

It has been shown above that the chemometric and graphical profile methods faithfully represent the Marlboro brand in specific geographical regions over defined time periods. Parallel studies on several other major transnational brands indicate similar behaviour to this Marlboro exemplar. Thus with careful calibration and monitoring trace element fingerprinting can provide an authentication tool independent of the tobacco industry and one that does not depend on packaging or security features being robust to counterfeiting efforts. The sources and proportions of each tobacco type in a given blend are normally confidential information so it is essentially impossible for counterfeiters to replicate the intrinsic trace element composition of tobacco because the process of adding some trace elements and extracting others to achieve a matching blend is not practicable. While many of the packs studied are quite old and were not selected to be representative of the present day illicit market it is noteworthy that substantial numbers appear to be indistinguishable from legal products, raising the question as to how they may have been diverted from the legal to the illicit supply chain and whether smuggling was involved in their distribution.

Trace elements can also contribute to reconstructing supply routes by matching seizures within or between jurisdictions and over intervals of time. Similarly provenance indicators can contribute to reconstruction efforts. The provenance examples presented (Fig.4) were significantly limited by the small size of the underlying chemometric database. A global collaborative effort to sample the main tobacco-growing countries could lead to a

substantially improved database and thus in all likelihood much better geographical resolution of provenance.

The trace element method is not suitable for empty (or discarded) pack surveys as little or no tobacco remains for analysis, however independent authenticity testing on packaging has recently been shown be practicable using ultraviolet radiation and microscopic examination of print quality.[33]

Industry conflicts

A case for independent testing has been stated above. The tobacco manufacturers regularly investigate large numbers of seizures to determine authenticity for statistical purposes. [10] Furthermore tobacco companies that signed agreements with the EU are regularly requested to test seizures to determine whether they are liable to restore evaded taxes associated with the illicit supply of their own products. In order to avoid conflicts of interest representative subsets (at least) should be sent routinely for independent analysis using a methodology not dependent on proprietary information. This and other recent work shows that independent methods are available to authenticate seizures without the conflicts of interest inherent in the present arrangements. [33] The manufacturers agreements with the EU are open to criticism for lack of transparency and failing to compel the manufacturers to secure their supply chains. [34] There is now a strong case for developing protocols and standards so that the formal role of product authentication can be carried out by organisations independent of the industry.

Implications for policy and practice

A review in 2010 estimated the global trade in illicit cigarettes as 11.6% of total products with 16.8% in low-income and 9.8% in high-income countries, resulting in a loss of about $40.5 billion a year in government revenues.[35] Excess mortality attributable to the availability of

cheap tobacco products affects middle- and low-income countries disproportionately. The FCTC policy of Track and Trace has a major role in reducing these impacts on health and inequalities however when an illicit product carries no T&T technology or fails the authenticity test it is not possible to extract any further information. The trace element method described above leads to a graphical "fingerprint" for an individual tobacco product and a statistical methodology for advising on its legality, while also identifying any matches and constraining the source of tobacco. Track and trace technologies and trace element fingerprinting thus address different aspects of the illicit product. They are complementary activities and for greatest effect both approaches should be integrated with conventional intelligence gathering in a single strategy for tackling the availability of illicit tobacco through disrupting supply chains. Implementing this strategy would benefit considerably from the establishment a global-scale repository of local tobacco leaf and products especially from regions that tend to supply the illicit manufacturers. These aims are consistent with Article 23 of the FCTC illicit trade protocol that requires parties to cooperate in expertise and technologies for forensic analysis and to develop methods for identifying the geographical origin of seized tobacco and tobacco products. [4]

Establishing a laboratory to apply trace element profiling has significant resource implications. The present work was conducted using laboratory space of about $10m^2$ housing an X-ray fluorescence spectrometer along with ancillary sample preparation equipment. Spectrometer calibration is a specialist task but thereafter tobacco samples can be prepared and run without need for highly specialised training assuming that quality assurance is embedded in the analytical protocol. Each sample takes about 10 minutes to prepare and requires up to 100 minutes of analysis time. With continuous sample feed for unattended analysis it is possible for one person to prepare and run single analyses of approximately 100 tobacco samples per week. Recent developments in X-ray analysis and other spectroscopic techniques could soon

lead to complementary new techniques for tobacco profiling that could be applied in field settings away from a central laboratory, potentially leading to screening methods with rapid response times and reduced resource needs.

Although this study has focused entirely on the Marlboro brand very similar results have been obtained for other major transnational brands despite fewer counterfeits being available for analysis. Trace element fingerprints have a generic usefulness that can be extended to other issues including understanding the complexities of cheap (illicit) white products that have come to dominate the illicit market in Europe.[7] FCTC's policy of Track and Trace is not a complete solution to the illicit tobacco problem and other policies for dealing with earlier stages in the supply chain of an increasingly diverse range of illicit products will be necessary if their supply is to be reduced significantly. [9] Some promising new approaches have been identified recently including microscopy, UV spectroscopy, radiogenic isotopes and DNA barcoding.[33 36 37] A programme to find their relative strengths and weaknesses and to develop improved methods and associated reference databases is warranted.

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FIGURE CAPTIONS

Figure 1. Discriminant function analysis using trace elements in tobacco from major EU brands classified by manufacturer. Tie lines join individual pack compositions to the discriminant function means for the manufacturer's brands in the dataset.

Figure 2. Trace element profiles for tobacco in packs of Marlboro legally purchased in the EU (red lines) and the US (blue lines). See text for details of profile construction. The field shaded yellow envelopes two standard deviations either side of the mean profile for legal EU Marlboro.

Figure 3. Trace element profiles for 35 packs of illicit Marlboro seized in the EU. 18 profiles green lines) mainly fall within the yellow field that defines 95% of EU Marlboro and are interpreted as authentic. 17 other profiles (red lines) mainly fall outside this field and are interpreted as counterfeit. Two of the counterfeits (S26 & S29) are shown separately to illustrate the similarity of their profiles. These are interpreted as having the same or closely related sources of tobacco.

Figure 4. Chemometric distribution of trace elements in 35 packs of illicit Marlboro seized in the EU plotted in terms of the first and third principal components. The data points are superimposed on the geographical distribution of local (non-transnational) brands implying a strong Asian affinity of most counterfeits (S19-35).
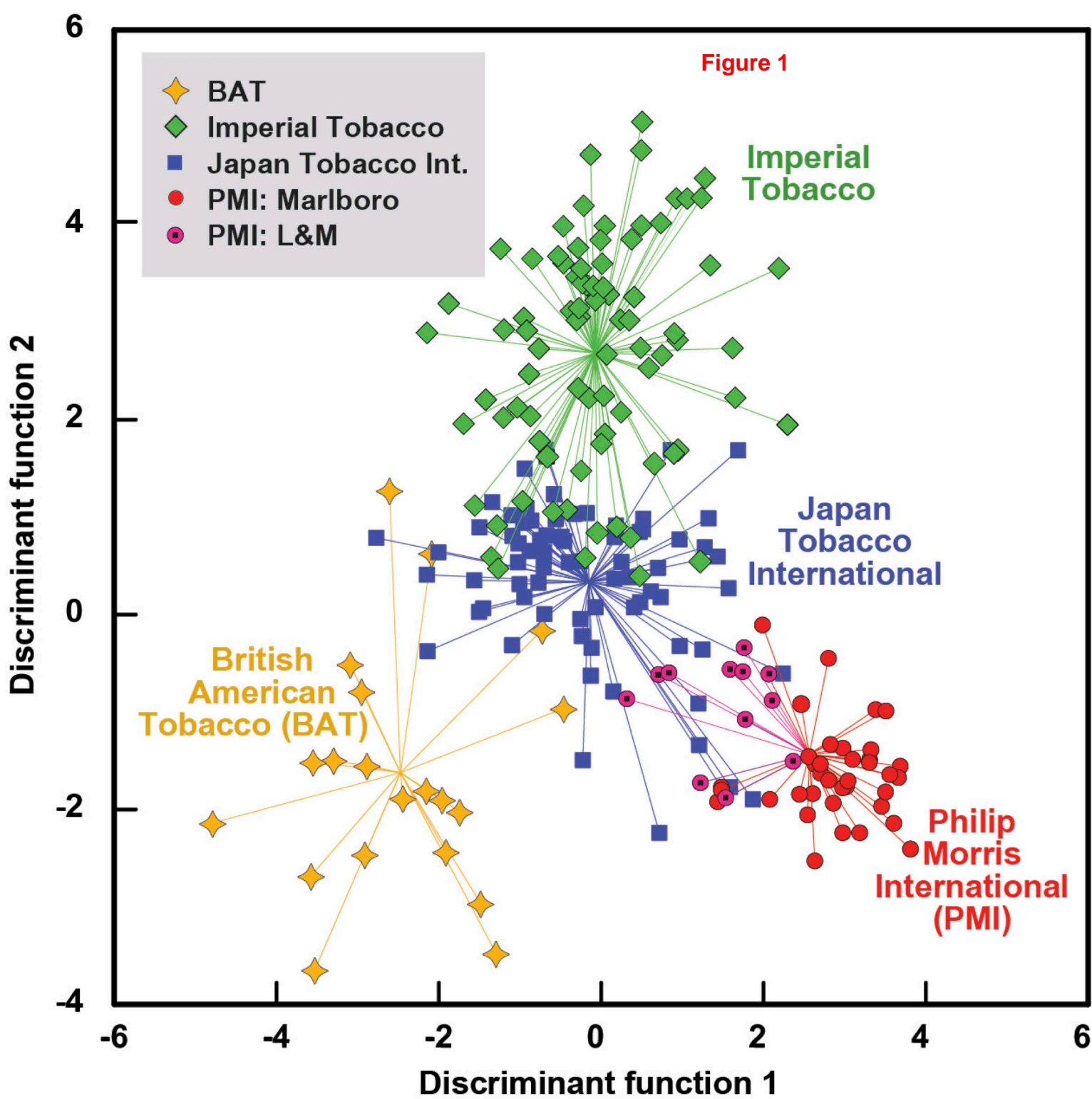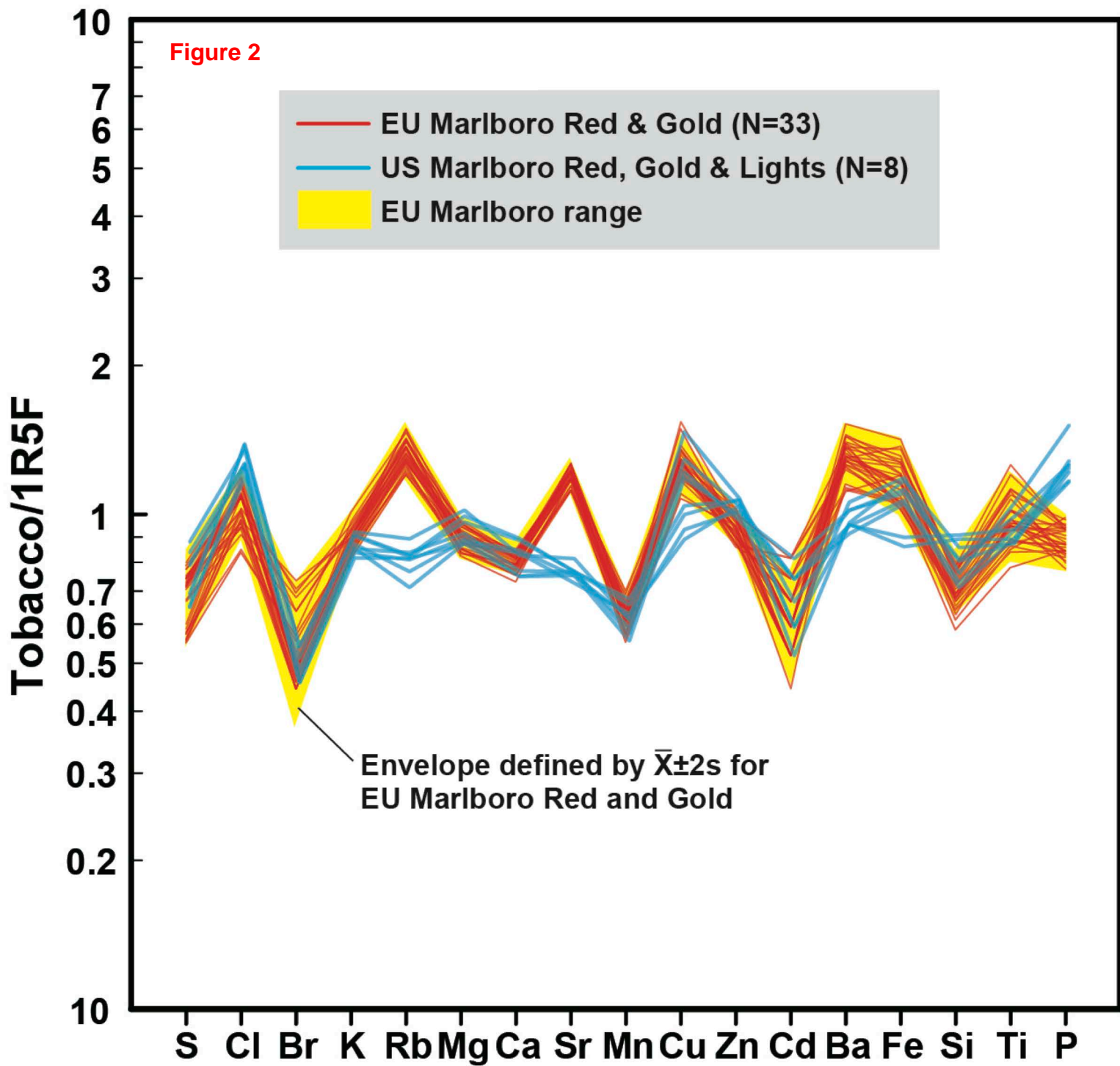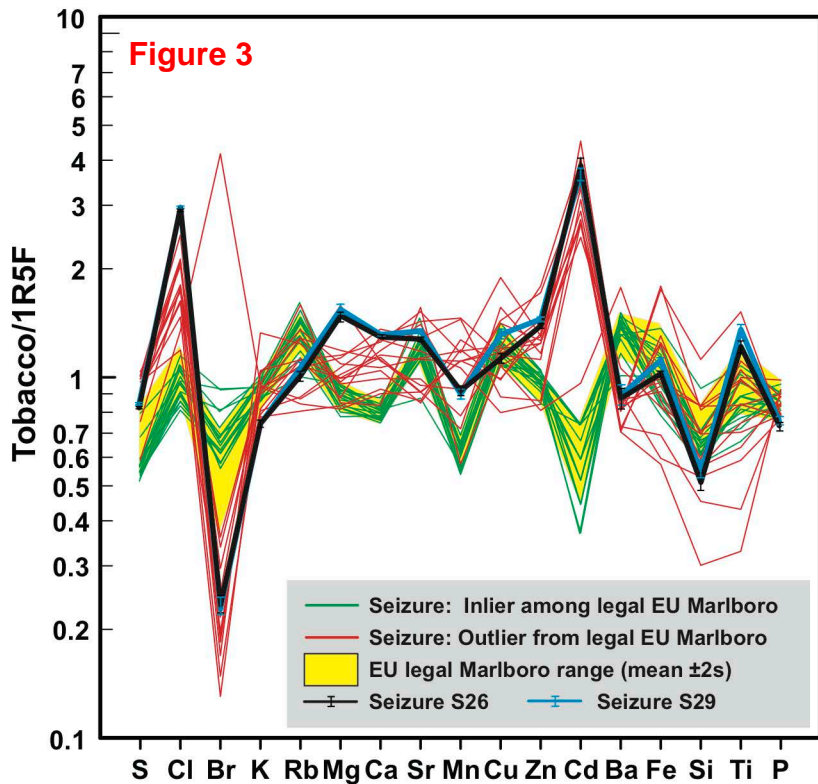
Figure 1

Figure 2

Figure 3

Figure 4