

FORENSIC INTELLIGENCE ON ILLICIT MARKETS:
THE PHYSICAL AND CHEMICAL ANALYSIS OF
WATCHES AND ITS CONTRIBUTION TO THE FIGHT
AGAINST COUNTERFEITING

PHD THESIS
SARAH HOCHHOLDINGER

ECOLE DES SCIENCES CRIMINELLES
UNIVERSITY OF LAUSANNE - SWITZERLAND

LAUSANNE, 2019

Supervisors:

Professor Olivier Delémont

Professor Pierre Esseiva

IMPRIMATUR

A l'issue de la soutenance de thèse, le Jury autorise l'impression de la thèse de Mme Sarah Hochholdinger, candidate au doctorat en science forensique, intitulée

« Forensic Intelligence on Illicit Markets: the Physical and Chemical Analysis of Watches and its Contribution to the Fight Against Counterfeiting »

Le Président du Jury

Professeur Andy Bécue

Lausanne, le 13 décembre 2019

Acknowledgements

Writing this thesis, from the initial choice of a topic, through various methodological challenges, data analysis and of course drafting the final manuscript was a long and winding road. Every aspect of this research brought new challenges. The light at the end is an overwhelming personal accomplishment beyond description. I would like to take this opportunity to thank all those whose assistance was a milestone in the completion of this project.

First of all, I wish to express my sincere appreciation to my supervisors, Professor Olivier Delémont and Professor Pierre Esseiva. I want to thank you for the confidence you had in me, for giving me so much freedom for my research, for your persistent help and guidance, as well as all your encouragements.

I particularly thank Michel Arnoux, Head of the Anti-Counterfeiting Department at the Federation of the Swiss Watch Industry (FH), for accepting this collaboration. I want to thank you for giving me the opportunity to do an internship at the FH, for all your knowledge you shared with me on counterfeit watches, for being a member of the jury, for your kindness and your great interest in forensic science.

I also wish to express my gratitude towards the Cantonal Laboratory of Vaud, in particular Dr. Stefan Bieri and Dr. Laure Marvin. Thank you for the pleasant time in your laboratory and your great help during the development of the analytical method.

I would like to thank Professor Quentin Rossy for being a member of the jury and the precious comments on my research.

I would also like to thank Dr. Carlos Fraga from the Pacific Northwest National Laboratory for taking the time to review my research.

Special recognition goes to the Director of the Ecole des sciences criminelles, Professor Olivier Ribaux and his team.

A big thank to the EDL team, Susanna, Valentin, Aline and Eric. It was always a great pleasure to work with you.

I want to thank my friends, Bettina, Fabian, Loretta, Lukas and Nicole for the unforgettable years of university studies.

I would like to thank my colleagues and friends Anne-Céline, Anne, Harmonie, Luca, Guillaume, Francesco, Sorcha, Mathieu, Saman, Killian, Robin, Daniel, Susanna and Natalia for all the moments we have shared together during the past five years. Your presence has been very important to me and you have always been a major source of support. A special thanks to Lydie and Lisa for your constant assistance, open ears, honesty and friendship.

I wish to acknowledge the support and love of my siblings, Patrick and Nina as well as my sister-in-law Tamara.

Words cannot express the gratitude I feel towards my parents, Jürg and Monika. You gave me the strength to reach for the stars. I deeply honor your selfless love, care, pain and sacrifices that you have made throughout my life.

I wish to express my deepest gratitude to my boyfriend Men. Thank you for your belief in me and for standing beside me with all your love and endless support.

Abstract

Counterfeit luxury fashion goods have rarely been the subject of scientific studies. Very little is known about the mechanisms of this illicit market, despite the apparent prevalence and their adverse consequences. Contrary to the general perception of counterfeiting having minimal real-world consequences, it can indeed have serious effects on the economic, social and human levels. The profits derived from the industry are largely associated with revitalising other forms of underground economies and illicit activities. In the luxury goods segment, watches remain one of the preferred targets of counterfeiters. Very little is known on the production of these watches and the relationships among the different steps of raw material supplies, watch part manufacturing, assembling and diffusion.

The study of traces in a forensic intelligence perspective can contribute to an improved understanding of the phenomenon. The aim of this research was to highlight different types of links that can be drawn between specimens of counterfeit watches, to carry out a thorough study of the information conveyed by the revealed links, to study their complementarity and to get an understanding of the knowledge that can be produced from these pieces of information. A further and complementary objective of this research was to appraise the potential risk to public health due to toxic heavy metals in watchcases and its relevance as an anticounterfeiting approach.

The first part of this research was devoted to the development of a suitable analytical strategy providing representative and informative data on the composition of watchcases and the creation of reliable profiles from these analytical results. Subsequently, these profiles were compared and grouped. In the second part, a thorough study of physical features as well as the relationships within the analysed watches was performed.

Chemical and physical links were found that corroborated existing knowledge and also revealed new connections between different seizures or specimens. The comparison with spatiotemporal information elucidated possible aspects of the structure and the organisation of production and distribution channels.

The detection of heavy metal contaminations demonstrated that some counterfeit watchcases are manufactured with inferior materials. These results presented a more comprehensive understanding on the effects of counterfeiting; showing that they are not just restricted to intellectual property and trade infringements but may also pose a public health problem.

The proposed approach, oriented from a forensic intelligence perspective, allowed to combine different dimensions in an interdisciplinary manner. The resulting insights provided valuable investigative leads, a solid basis for future research and demonstrated how forensic science can actively contribute to the gathering of intelligence on illicit markets.

Résumé

La contrefaçon de produits de luxe a rarement fait l'objet d'études scientifiques. Les mécanismes de ce type de marché illicite restent très peu connus, malgré l'apparente prévalence et les effets négatifs de celui-ci. En effet, malgré la perception générale que les conséquences liées à la contrefaçon seraient négligeables, le marché de la contrefaçon peut, en fait, engendrer des effets sérieux au niveau économique, sociale et humain. Les bénéfices tirés de cette industrie sont largement associés au financement d'autres formes d'économies clandestines et activités illicites. Dans le secteur du luxe, les montres demeurent la cible privilégiée des contrefacteurs. Il existe très peu d'informations quant à la production de ces montres et les relations qui existent entre les différentes étapes de l'approvisionnement en matières premières, la fabrication des composants de montres, l'assemblage et la diffusion.

L'étude de traces dans une perspective de renseignement forensique peut contribuer à une meilleure compréhension du phénomène. Les objectifs principaux de cette recherche étaient de mettre en évidence les liens qui peuvent être établis entre des spécimens de montres contrefaites, puis de procéder à une étude exhaustive des informations obtenues à partir de ces liens, d'étudier leur complémentarité et enfin, de comprendre les connaissances qui peuvent être tirées à partir de ces différents éléments d'information. Un autre objectif complémentaire était d'évaluer le risque potentiel pour la santé public de la présence de métaux lourds dans les montres contrefaites et la pertinence de cette observation en tant qu'approche de lutte contre la contrefaçon.

La première partie de cette recherche était consacrée au développement d'une stratégie analytique adaptée à la production de données représentatives et informatives sur la composition des boîtiers de montres et à la création de profils fiables à partir des résultats analytiques. Par la suite ces profils ont été comparés et groupés. Dans une deuxième partie, une étude approfondie des caractéristiques physiques et les relations qui existent entre les spécimens analysés a été effectuée.

Les liens chimiques et physiques établis ont corroboré des connaissances existantes mais ils ont également révélé de nouvelles connections entre saisies et spécimens. La comparaison avec des informations spatiotemporelles a mis en évidence des aspects

possibles de la structure et de l'organisation de la production et des canaux de distribution.

La présence de contaminations de métaux lourds a démontré qu'une partie des boîtiers de montres contrefaites sont fabriquées avec des matériaux de qualité inférieure. Ces résultats ont permis de parvenir à une meilleure compréhension des effets de la contrefaçon en montrant qu'elle n'est pas limitée à une atteinte aux droits de propriété intellectuelle mais qu'elle peut également poser des problèmes de santé publique.

L'approche proposée combine plusieurs dimensions d'une manière interdisciplinaire. Les résultats obtenus ont permis de fournir des pistes utiles, une base solide pour des recherches futures et ils ont démontré que la science forensique peut activement contribuer à la production de renseignement sur les marchés illicites.

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1 Introduction

1.1 Definition of counterfeiting

Counterfeiting and piracy are notions used to describe the violation of Intellectual Property Rights (IPRs), but currently no broadly accepted definition exists. In general, the term “counterfeiting” refers to violations of trademarks, design rights and patents, whereas “piracy” refers to copyright infringements [1]. Counterfeiting takes into account physical products and brands while piracy considers the reproduction of intellectual content products (such as movies, books, music and software) [2].

In this study, we focus on “fake” watches, which fall within the category of counterfeits. We refer to “counterfeiting” as “*...unauthorized copying of an original product – counterfeiters try to imitate the characteristics, materials or look of an original or legally protected product in order to take advantage of the reputation of the original, which is based on a trademark, an indication of source (such as "Swiss Made"), a design or a patent*”, a definition stated by ‘STOP-PIRACY’¹, the Swiss platform against counterfeiting and piracy.

Even though counterfeiting is a form of illicit trade, it is important to provide an overview of other varieties of illegal market activities. They differ considerably in their enablers, driving factors, actors and impacts they cause on affected institutions. In the same sense, counterfeiting and piracy are often lumped together in publications but are clearly different when it comes to analysing the phenomenon or proposing effective countermeasures. The overview presented in Figure 1 proposes a subdivision of illicit trade activities and refines IPR infringements up to counterfeit physical goods. It is generally accepted to distinguish physical counterfeit goods into two categories as suggested by Grossman and Shapiro in 1988: deceptive and non-deceptive goods [3]. Buyers may acquire counterfeit goods by not being aware that it is a “fake” (deceptive good). The problem of these goods is that they are extremely competitive with the original goods. On the other hand, for non-deceptive good, buyers may have reasonable

¹ Retrieved from the website STOP-PIRACY: <http://www.stop-piracy.ch/gefalscht-und-kopiert!/definitionen/> (last accessed: 02.03.2020)

doubts regarding authenticity. In particular in the area of counterfeit luxury goods, it was shown, that the majority of people buy them voluntarily and know that it is a counterfeit, mostly based on the price, sales channel and inferior quality of the product [4]. This dichotomy might not always be clear since a wide range of quality, price and deception among counterfeit products exist [5].

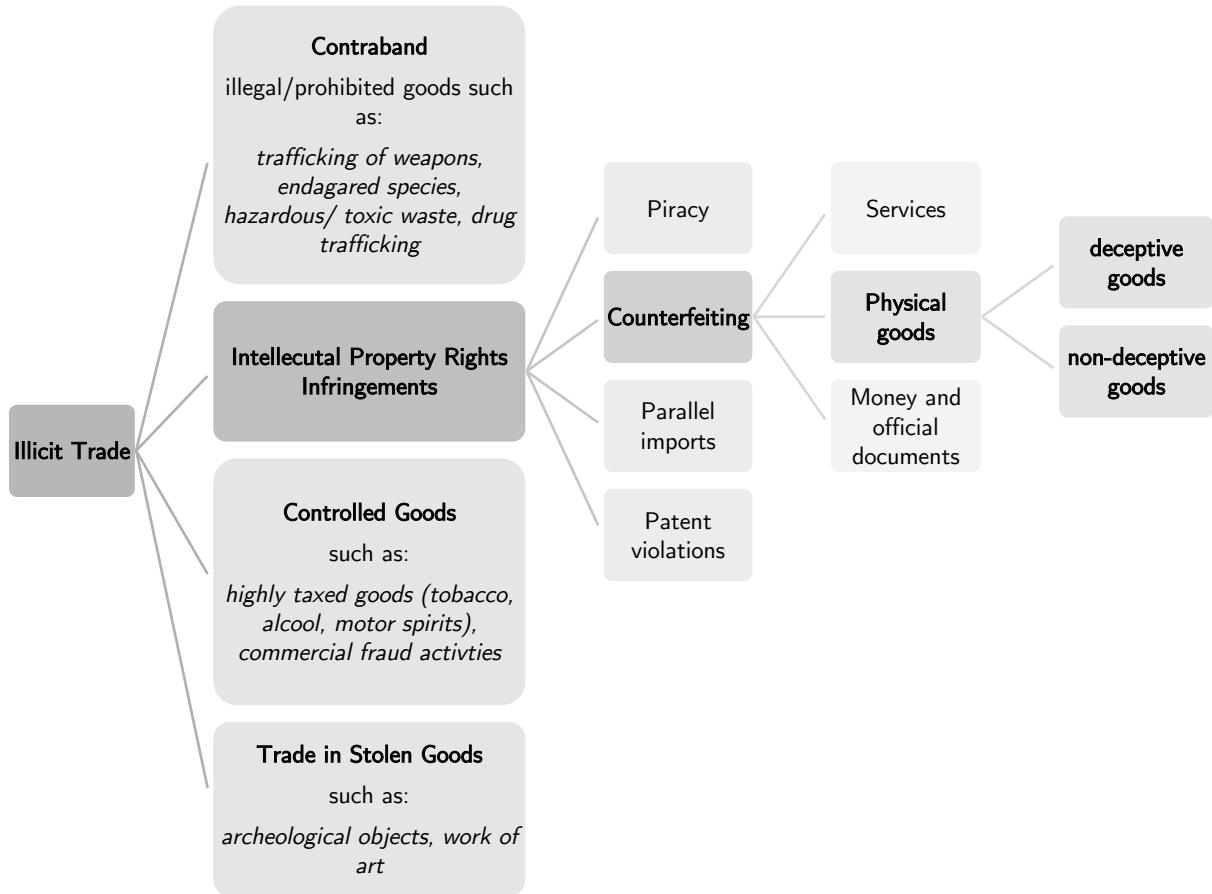


Figure 1: Overview of illicit trade activities and integration of counterfeiting and its subdivisions, adapted from Staake et al. [6] and WCO 2017 Illicit Trade Report [7]

1.2 Counterfeiting: a brief overview

1.2.1 Statistics

Counterfeiting is said to be the second oldest profession in the world. It is also one of the most lucrative businesses. Since counterfeiting is a clandestine activity, it is practically impossible to recover accurate statistics, in particular due to difficulties in measuring production and sales [8].

Moreover, it has to be kept in mind that existing statistics often present numbers of interventions without however providing the actual number of seized goods. They reflect more the strategies that custom authorities and law enforcement institutions put into a specific product sector over a certain period of time. Numbers between the different attempts measuring the counterfeit market vary greatly. This is mainly due to the lack of consistent definitions and categorizations of the concerned goods as well as challenging measurement methodologies, with no agreement on accounting factors [9]. No methodology exists to accurately measure the overall magnitude of counterfeit products [10, 11]. Consequently, only approximate estimates can be drawn from information collected by customs, law enforcement, investigative agencies and surveys. The ‘Organisation for Economic Co-operation and Development’ (OECD) estimated the trade in counterfeit goods up to USD 200 billion in 2005 [11]. According to data based for 2013, the international trade in “fakes” even accounted for USD 461 billion, representing as much as 2.5% of the total international trade [1]. An updated estimation of the global value of counterfeit and pirated products, based on international trade statistics and customs seizures² in 2016, even reached USD 509 billion [12]. This represents 3.3% of world trade. The share of trade in counterfeit and pirated goods seems to grow significantly. This can be partly explained by a reduction in volumes of world merchandise trade between 2014 and 2016, dramatic increase of small parcels crossing borders and lightly regulated free trade zones (FTZs) [12]. Asia is found to be the greatest production site (and China and Hong Kong the most important source economies) for counterfeit products [1, 11, 12].

Collated data from customs, law enforcement institutions, industry and research organisations suggest, that the type of products being counterfeited are abundant and diversifying. Counterfeiting not only refers to luxury and high-end products, but also to virtually all types of consumer goods (such as comestible goods, tobacco, automotive parts, personal-care products, and toys) [9]. In a general perception, counterfeiting is an increasing phenomenon. Nonetheless, available statistics fail to adequately validate these perceptions, since counterfeits are produced and sold outside any recorded market

² The database on customs seizures was provided by the World Customs Organization (WCO) and complemented with regional data submitted by the European Commission’s Directorate-General for Taxation and Customs Union, the US Customs and Border Protection Agency and the US Immigration and Customs Enforcement.

[8]. Like any other clandestine activity, estimations on scope and magnitude rely on statistical data that are largely incomplete. Vagg and Harris also identified that counterfeiting is given a rather low priority by law enforcement agencies [13]. Hence, it is likely that the major part of counterfeit trade remains undetected. Any claims about scope and scale of counterfeit trade should therefore be critically questioned [14].

1.2.2 Negative effects

The negative consequences of counterfeiting are perceptible in many ways. They have an effect on a socio-economic level, on governments, right-holding businesses and consumers.

General socio-economic effects include a shift of employment from legal to illegal businesses. Negative environmental consequences may arise from the production of substandard products or from the disposal of seized counterfeit goods [11]. In 2015, for example, the ‘European Taxation and Custom Union’ destroyed over 15 million articles [15]. Furthermore, several reports and researches emphasize the fact that counterfeiting generates profits that help to finance other forms of crime [8, 15-22].

On a governmental level, the trade in counterfeit goods affects the host country of right-holding businesses, the country where counterfeiting takes place and the country where the counterfeits are sold. The host countries of the right-holding firms lose exports, taxes and employment [9]. Countries where counterfeits are produced and/or sold also experience tax revenue losses, growth in underground economy at substandard conditions (including child labour), reduction of legitimate employment, additional costs for anticounterfeiting operations and corruption [8, 9, 11].

Right-holders suffer from financial loss due to sales reduction and increasing costs for protecting and enforcing their IPRs. Frequently counterfeit brands risk damage to their reputation and brand value, possibly resulting in decreasing customer loyalty [9].

Impacts on consumers can be as harmless as the loss of a small amount of money and/or dissatisfaction of the purchased goods [9]. However, serious health and safety effects may arise due to the use of substandard products. Counterfeit pharmaceuticals are on that note of the greatest concern. Regarding public health and safety issues several other cases have been reported, including fake auto and aviation parts, toys, foods and beverages including counterfeit vodka and infant formula and various

consumer goods such as electrical cords that busted into flames during normal use, overheating batteries and shampoos with unsafe level of bacteria [18].

1.2.3 Driving factors

Why is counterfeiting such an attractive business? According to estimates on the revenue of illicit markets in Europe, counterfeiting generates 43 billion Euros per year compared to 28 billion Euros for trafficking in illicit drugs (heroin, cocaine, cannabis, and amphetamine drugs) [19]. These figures clearly demonstrate the tremendous profits that can be gained by selling counterfeit goods. In addition to the large pay-outs, conventional costs for research, developing, meeting health and safety standards, quality control and advertisement are avoided. Consumers play their part by willingly purchase the counterfeit goods, partly due to superficial exclusivity and the association with prestige [23].

Counterfeit goods are mainly sold via informal markets and the Internet. The OECD identified the main reasons for the attractiveness and expansion of the Internet as a sales platform for counterfeit goods, as resumed in Table 1 [11].

| | |
|---------------------|--|
| Anonymity | Diminishes the risk of detection |
| Flexibility | Rapid, simple and inexpensive creation of websites |
| Market Size | The large number of e-commerce sites make counteractive measures difficult |
| Market Reach | Global market reach, available 24/7 |
| Deception | Simple creation of confusingly similar looking websites |

Table 1: Driving factors for counterfeiters' attraction to the Internet

Law enforcement on a national and international level is weak and, in some countries, even non-existent. Penalties are often very low and therefore non-dissuasive and rarely enforced [24]. An additional challenge for international cooperation for law enforcement is the local, national and global cross-linking of counterfeiting chains found in societies with different legal systems, cultures, values and priorities [25].

1.2.4 Protection of intellectual property rights – legal aspects

Governments and industries strive to combat counterfeiting. On a global level, several intergovernmental institutions, such as Interpol, the ‘World Customs Organization’ (WCO) and the ‘World Intellectual Property Organization’ (WIPO) advise and assist different initiatives and provide training and technical support for governments. Basic multilateral rules are specified in the Agreement on ‘Trade-Related Aspects of Intellectual Property Rights’ (TRIPS) issued by the ‘World Trade Organization’ (WTO).

Alongside these global institutions, there are national initiatives. In Switzerland, governmental agencies involved in the protection of IPRs are the Federal Customs Administration, the Federal Office for Police, the State Secretariat for Economic Affairs (SECO) and the Federal Office of Consumer Affairs. The registration office for property rights is the Swiss Federal Institute of Intellectual Property [11].

The legal basis for the protection of IPRs is to be found in the ‘Federal Law on the Protection of Trademarks and Indications of Source’ (RS 232.11), the ‘Federal Act on the Protection of Designs’ (RS 232.12) and the ‘Federal Law on Patents for Inventions’ (RS 232.14). Manufacturing, selling and importing counterfeit goods into Switzerland is prohibited. Custom authorities can withhold and destroy counterfeit goods, from both, personal luggage or parcels, that arrive or transit by mail³. Educational and awareness campaigns are organized by the association ‘STOP PIRACY’ in cooperation with the public and private sector⁴.

1.2.5 A non-conventional crime

The difficulty to accurately quantify counterfeiting was clearly demonstrated. Moreover, very little is known about the structures and mechanism of these illicit markets and even less is known on the market of counterfeit luxury fashion goods.

³ Information retrieved from the website of the Swiss Federal Institute of Intellectual Property: <https://www.ige.ch/en/intellectual-property/counterfeiting-and-piracy/what-does-the-law-say.html> (last accessed: 02.03.2020)

⁴ Information retrieved from the website of STOP PIRACY:
http://www.stop-piracy.ch/StopPiracy/media/_content/PDF/c101e.pdf (last accessed: 02.03.2020)

Furthermore, different public and private interests are involved compared to safety-critical goods, such as counterfeit pharmaceuticals [25].

Counterfeiting is an atypical phenomenon since it differs from the traditional form of crime [14]. First of all, it is not possible to clearly define who the victim(s) of counterfeiting is/are, but possible distinctions can be made [25]:

1. Consumers
2. Affected brands
3. Indirectly affected victims (e.g. environment, taxation system)

Naturally, the resulting question refers to the perpetrator – consumer and/or counterfeiter? A distinction could be made considering if the product is deceptive or not. But then, how could it be proven if a consumer knowingly purchased a counterfeit good. Hence it is difficult to criminalize possession of counterfeit goods, although some countries have a zero-tolerance policy.

Inevitably, there is a confusion among public and private interest to combat counterfeiting. Where are the responsibilities of consumers, of brands and of governments or – put another way – should the taxpayer or the IP owners finance anticounterfeiting measures [25]. Obviously, the interests of fighting IPR infringements in industrialised and developing countries are also contradictory and may even raise questions from an ethical perspective.

1.3 Watch counterfeiting

1.3.1 Scale of the phenomenon

The luxury goods industry, and especially the watch industry, is greatly affected by the problem of counterfeiting. The economic losses due to this phenomenon are estimated at about CHF 800 million for a total of nearly 40 million counterfeited watches offered for sale each year [26]. However, an explication how this number was obtained is not provided.

As mentioned above, it is extremely difficult to obtain accurate statistics for counterfeit products. Custom authorities provide certainly incomplete, but at least real data on seizures of counterfeit goods.

On a global scale, the WCO drafts the ‘Illicit Trade Report’ every year. A worldwide database, called ‘Customs Enforcement Network’ (CEN) contains customs seizures and offences and is filled by the WCO members (customs authorities).

In 2013, 1'733'450 counterfeit watches were introduced into the CEN database [27], 1'411'587 in 2014 [28] and 1'289'110 in 2015 [29]. From 2016 on, the data was reported in bar charts and no actual numbers were available anymore. In 2013 and 2015, Rolex held the 3rd respectively 11th place in the list of most counterfeited brands in terms of number of cases⁵. It has to be kept in mind that the CEN database excludes small-scale seizures and a database entry is only made for seizures with larger numbers of items or of high value. Furthermore, the data is gathered on a voluntary basis and reflects to some extent dedicated actions that were promoted by the WCO in some developing countries [1].

The ‘European Commission’ annually publishes a report on EU customs enforcement of intellectual property rights. Table 2 shows a breakdown for the watch sector between 2012 and 2018.

| Year | Number of interceptions by customs (watch sector) | Number of seized counterfeit watches | Retail value of original goods [€] |
|------|---|--------------------------------------|------------------------------------|
| 2012 | 7'376 | 471'366 | 177'176'529 |
| 2013 | 4'942 | 324'293 | 158'325'350 |
| 2014 | 4'597 | 182'817 | 92'163'973 |
| 2015 | 5'972 | 198'202 | 154'251'462 |
| 2016 | 5'067 | 198'804 | 109'754'626 |
| 2017 | 5'621 | 207'604 | 171'485'023 |
| 2018 | 4'922 | 91'271 | 117'156'752 |

Table 2: Numbers of interceptions by customs, numbers of seized counterfeit watches and retail value of original goods. The numbers represent customs actions at the EU external borders [15, 30-35].

In Switzerland, the ‘Federal Customs Administration’ (FCA) publishes the ‘Intellectual Property Law Statistics’ [36-42] every year. Numbers of interventions for jewellery and watches between 2012 and 2018 are shown in Table 3.

⁵ For the other years, the top counterfeited brands were not mentioned.

| Year | Total number of interceptions by customs | Interventions for jewellery and watches ⁶ [%] |
|------|--|--|
| 2012 | 2'741 | 6.4% |
| 2013 | 2'764 | 8% |
| 2014 | 2'856 | 9% |
| 2015 | 3'621 | 30.2% |
| 2016 | 3'125 | 49% |
| 2017 | 1'633 | 36% |
| 2018 | 1'686 | 37.4% |

Table 3: Intellectual Property Law Statistics from the Federal Customs Administration (FCA)

Some efforts were made trying to improve estimating the magnitude of counterfeit trade. Measures are principally based on international trade and customs seizures data. Reports issued by the OECD calculated the ‘General Trade-Related Index of Counterfeiting’ (GTRIC) for products (GTRIC-p) and provenance economies (GTRIC-e). The methodology is based on seizure intensities in each product category weighted by the share of general trade volumes of each reporting economy for these products. Accordingly, it is possible to appraise the extent of counterfeiting within a specific product sector and the overall magnitude [1, 12].

Table 4 shows that watches are one of the product categories most likely to be counterfeited. As also stated in other reports [11, 17], Table 5 shows that Hong Kong and China are the most cited source economies.

| Top 5 product categories | GTRIC-p |
|--------------------------------|---------|
| Watches | 1.000 |
| Articles of leather, handbags | 1.000 |
| Clothing, knitted or crocheted | 1.000 |
| Perfumery and cosmetics | 1.000 |
| Footwear | 1.000 |

Table 4: GTRIC-p values for the top 5 product categories. GTRIC-p is the relative probability for product categories to contain counterfeit goods [12]. The values are based on average data from 2014-2016.

⁶ Proportion of the total interventions by categories of goods

| Top 5 provenance countries | GTRIC-e |
|----------------------------|---------|
| Hong Kong (China) | 1.000 |
| China | 1.000 |
| United Arab Emirates | 0.995 |
| Morocco | 0.989 |
| Pakistan | 0.955 |

Table 5: GTRIC-e values for the top 5 provenance countries. GTRIC-e is the relative probability for provenance countries to contain counterfeit goods [12]. The values are based on average data from 2014-2016.

It has been argued that EUR 1.9 billion of revenue are annually lost in the jewellery and watch sector in the EU marketplace (not including Switzerland) leading to 28,500 losses of employment and EUR 600 millions of losses in government revenues [43]. These numbers are based on a model including economic variables and variables related to counterfeiting. However, the methodology as well as the origin of the data is difficult to understand.

It has to be kept in mind, that statistics on counterfeit watches often lack in transparency and there is a constant risk of misreading or misinterpreting the numbers. It is virtually impossible to identify the key reasons for a drastic increase or decrease of intercepted counterfeit watches between two consecutive years. A real increase in production or sales is undetectable because of the clandestine nature of these activities. Custom authorities may intercept counterfeit watches in a more effective way (e.g. through training of customs officials). Furthermore, it is possible that customs authorities specifically target strategically counterfeit watches over a certain period of time.

1.3.2 Nature of the problem

None of the previously cited reports on counterfeiting specifies details on the seized watches. Very little is known on specific brands and models. A study carried out at the ‘Ecole des Sciences Criminelles’ (ESC) in 2016 aimed at collecting intelligence on the counterfeiting market through the study of Internet forums. It was possible to compile interesting statistics on the most discussed brands and models. Three main discussion forums were analysed by automatically extracting keywords from discussions. It was found, that the most popular brands were Breitling, Hublot, IWC, Omega, Panerai,

Rolex and Tag Heuer (not necessarily in this order). Concerning the models: Seamaster (Omega), GMT_Master (Rolex) and Cosmograph Daytona (Rolex) took the first places [44]. Internet forums constitute an open source of data that can be useful to assess popularity, tendencies and evolution of an online illicit market. The Federation of the Swiss Watch Industry (FH) also holds internal statistics on brands and models at risk, in order to elaborate efficient strategies for custom authorities. These statistics indicate that the counterfeiters generally target only two models of a brand [45].

1.3.3 Possible impacts and risks associated with watch counterfeiting

Most of the negative consequences of counterfeiting (as explained in section 1.2.2) also relate to watch counterfeiting.

First of all, there are costs to the right-holders in terms of losses of sale. Direct financial loss is impossible to determine. Some economists even claim there are no losses, since the buyer of a counterfeit good would not buy the genuine product [9]. This argument does not seem convincing in every case, since high-quality “fakes” partly compete with the original goods [8]. On the other hand, it is certainly wrong to equate the loss of sales with the retail value of the goods, if they were genuine.

In addition to the negative financial impact, counterfeiting also affects brand value and reputation. Counterfeiting was identified as one of the top 5 risks in outlook for sales (among weaker foreign/domestic demand, smartwatches as a competitive threat and the strength of the Swiss Franc), in a study on the Swiss Watch Industry [46].

Some connections between watch counterfeiting and organized crime groups were already drawn. In 2002, for example, during a drug raid in a New York residence, 5000 fake Rolex watches were found (among other counterfeits). The prosecutor stated that the sales profits were used to launder drug money [18].

Counterfeit watches can also be dangerous to consumers. This refers to potentially hazardous compounds. The legislation in the European Union and Switzerland regulates the use and the amount of lead, cadmium, nickel, mercury and other toxic materials in consumer goods. Counterfeit goods are not subjected to these regulations and can potentially pose a hazard to consumers’ health.

1.3.4 Current anticounterfeiting approaches

One of the key actors in the fight against watch counterfeiting is the ‘Federation of the Swiss Watch Industry’ (FH). Currently, the organization represents around 500 members, accounting for approximately 90% of the Swiss watch industry manufacturers. Along with various tasks in developing and defending the sector, they actively fight IPR infringements. Their activities include analysis of fake watches, protection of Swiss geographical indications, market investigations, legal defence of intellectual property rights, awareness rising among authorities and the population and efforts to prevent the sale of counterfeit watches on the Internet.

In 2017, the FH dealt with over 2'500 cases in different countries around the globe, whereby over 1'900'000 watches were seized. In total, 397 watches, seized in 14 countries and violating 38 brands, underwent technical analysis. The vast majority of these watches originate from Asia, especially from China or in transit via Hong Kong [47].

The FH also draws particular attention to the evolution of the sale of counterfeit watches on the Internet. In 2015, the Internet Unit of the FH took down over 600'000 advertisements on platforms and social networks. They observed for the first time that the growth rate for new websites dedicated to sell counterfeit watches decelerated. The flip side of the coin is, however, a shift towards social media, accounting for 65% of the withdrawn ads in 2015 [45].

For many years, the ‘Ecole des Sciences Criminelles’ (ESC) of the University of Lausanne has been collaborating closely with the FH, which resulted in several master theses (c.f. section 2.2.2).

2 State of research

2.1 Research on counterfeiting

Some aspects of counterfeiting have been extensively studied. Major contributions can be found in the fields of marketing, law, management, psychology, business ethics and economics [48]. The different categories of research can be shown along the supply chain of both, licit and illicit trade, as shown in Figure 2.

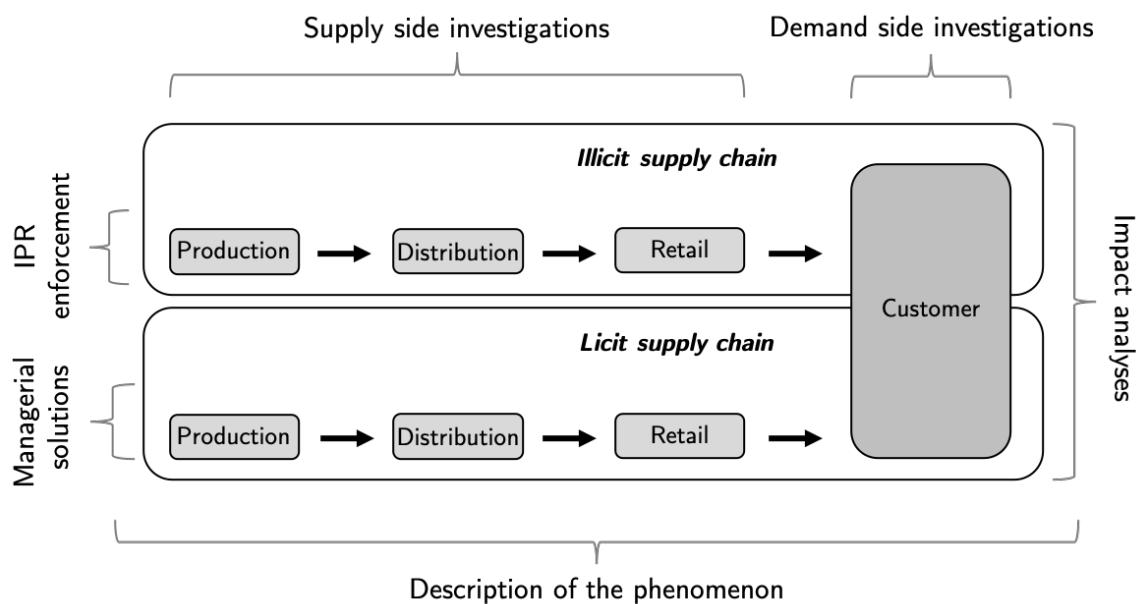


Figure 2: Areas of research on counterfeiting along the licit and illicit supply chain, adapted from Cesareo et al. [48] and Staake et al. [6]

Most scientific contributions in scholarly papers can be found regarding demand-side research [6]. This led to a solid understanding of consumer behaviour [48] in terms of who buys counterfeits and why they are bought. Furthermore, research on intellectual property rights is also comprehensive. Managerial solutions focus on corporate actions to combat counterfeiting, such as organisational and technical measures, legal protection and awareness raising among the public [49]. Supply side investigations are sparse although they are of great importance to understand the phenomenon's source and the operational structures of illicit markets. The main obstacle is, however, to gain access to market information [6]. Impact analyses try to assess the financial impact of

counterfeiting whereby methodologies lack amongst others in clear definitions, reliable data sources, sizes of units and assumptions on prices of counterfeit goods [50]. General descriptions on counterfeiting are mostly found in reports emitted by industry alliances⁷, governments⁸, various other institutions and organisations⁹ as well as in the press.

Nevertheless, counterfeiting, despite its apparent prevalence and negative consequences remains an under-explored area that needs further research. Possible explanations for this neglect could be the perception that IP crime is ‘victimless’ [14] and it is sometimes even valorised and justified as a disagreement and act of resistance against the market dominance of large corporations [51].

Staake et al. identified the problem that countermeasures are mainly based on observations of existing anticounterfeiting strategies although much could be directly derived from the characteristics of the counterfeit market [6]. Developing appropriate countermeasures requires the understanding of the phenomenon from various points of view. In this perspective, the study of the goods themselves originating from counterfeit markets may help to fill some of the knowledge gaps.

⁷ e.g. International AntiCounterfeiting Coalition, Anti-Counterfeiting Group, Union des Fabricants, Federation of the Swiss Watch Industry

⁸ e.g. Annual reports: “Report on EU customs enforcement of intellectual property rights” (European Union), “Intellectual Property Rights Seizures Statistics” (USA), “Intellectual Property Law Statistics from the FCA” (Switzerland)

⁹ e.g. World Trade Organization (WTO), World Customs Organization (WCO), Interpol, Organisation for Economic Co-operation and Development (OECD)

2.2 Intelligence drawn from the study of goods originating from illicit markets

2.2.1 Forensic intelligence as a methodological paradigm

The fundamental unit of forensic science is the trace. A physical¹⁰ trace can be defined as “pattern, a signal or material transferred during an event. It is the remnant (the memory) of a source and/or an activity that produced it” [52]. ‘Traces’ in their own sense are insignificant. If they are detected and identified as relevant for investigative purposes, they become ‘signs’. Through analysis and interpretation, forensic scientists seek to reveal relationships between people, places and objects in conjunction with the past action [53]. Signs eventually become ‘evidence’, if a signification is attributed as an explanatory element of knowledge to the past event [52, 54]. Traces, but also their evaluation, can take a variety of forms as well as many ways of interpretation. Forensic science has an important role beyond its historical and traditional perspective of presenting evidence in court on a case-by-case basis.

The information content of traces can contribute to models where intelligence and crime analysis tend to support tactical, operational and strategic decisions and crime prevention [55]. In this regard, traces are used as ‘intelligence’ rather than evidence. The role of material traces within this broader contribution was formalised to understand the interaction between material traces and intelligence [56-58]. Forensic intelligence can be defined as “*the interpretation of forensic case data to support a variety of different decisions in the interconnected web of processes crossing each policing system*” [59] or as “*...the accurate, timely and useful product of logically processing (analysis of) forensic case data (information) for investigation and/or intelligence purposes*” [58].

¹⁰ A different definition may be used for ‘digital traces’. The ASTM defines ‘digital evidence’ as: “information of probative value that is stored or transmitted in binary form” (ASTM International. E2916-19e1 Standard Terminology for Digital and Multimedia Evidence Examination. West Conshohocken, PA; ASTM International, 2019. <https://www.astm.org/Standards/E2916.htm>, last accessed 02.03.2020).

A digital trace can be defined as: “any modification of the scene that is stored or transmitted in binary form, subsequently observed, resulting from the event of interest.”

The definition of the trace implies its material existence and detectability [54]. Traces offer a source of information based on objective, measurable and comparable data [60]. The observation of similar features within the traces allows us to hypothesize on relationships between these traces and their cause (source or criminal activity) [61]. Forensic case data should be considered as an important source of information when dealing with any type of illicit activities. The forensic intelligence process includes the (systematic) collection, comparison, detection and analysis of traces and information. Analysis of links between cases provides information on the magnitude and the extent but also on the development of the phenomenon under study [59]. This approach ultimately supports decision-making in terms of priorities of investigations and preventive measures to be implemented.

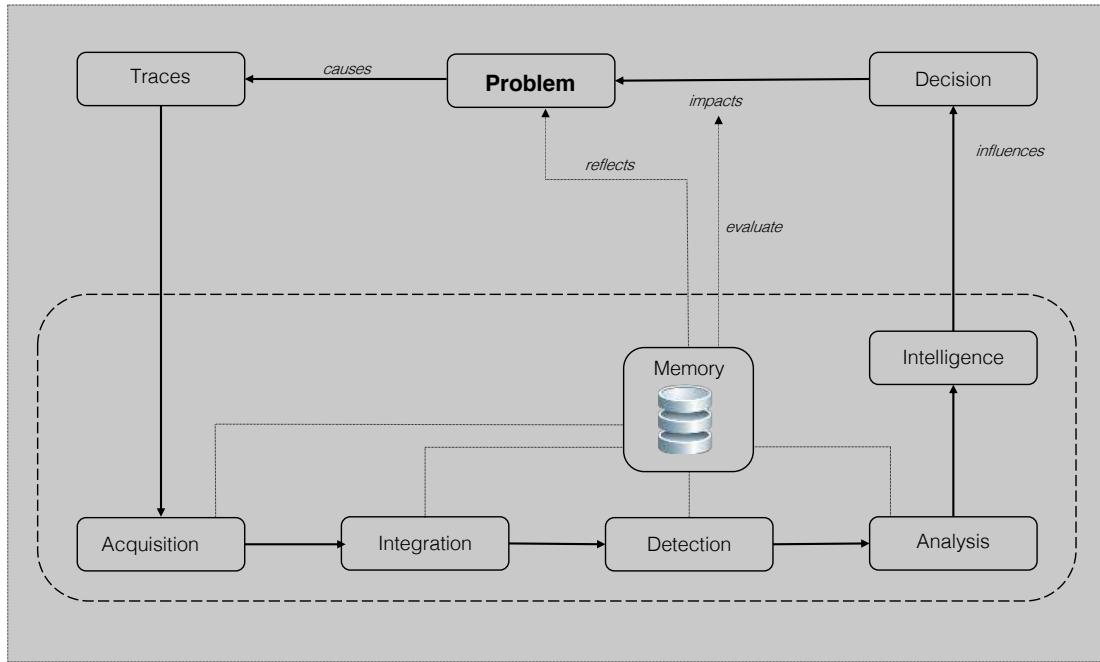


Figure 3: The forensic intelligence process, adapted from Morelato et al. [60] and Ribaux [62]

As shown in Figure 3, the process starts and ends with the problem itself. The resulting objects (e.g. a counterfeit watch, an illicit drug specimen or a false identity document) are one of the fundamental sources of the intelligence process. First of all, an object arising from the problem has to be detected. This step is obviously indispensable for both, traditional investigation and the forensic intelligence process. “*Its objective is to detect and recognise a concrete event or object arising from the problem, that is to say a physical remnant of criminal activity (i.e. trace). The aim is to distinguish events or*

objects related to the problem from those that are unrelated or legitimate. In general, one would consider this as a new case” [61].

The object under study is then examined and the relevant characteristics are extracted during the **acquisition** step. These characteristics can be of physical or chemical nature and are subsequently codified. The codified characteristics are called the ‘profile’ of the object.

Integration then covers the systematic organisation and classification of the acquired profiles. This operation aims at linking objects from a common source and separating them from traces of different sources. A link can be defined as an established relationship between two entities (e.g. profiles). The degree of relationship between profiles can be measured and expressed as a score. Depending on the comparison metric, scores may take the form of distances, correlations, similarity/dissimilarity or proximity measures [63]. Relationships between more than two entities are often referred to as ‘series’, ‘group’, ‘class’ or ‘cluster’. Repetitions are **detected** through the search for tendencies, clusters or elementary links and grouped into new problems [64]. Such specific configurations are called ‘patterns’. A pattern refers to a set of relations reflecting repetitive crime behaviour within the collected data [65].

The **memory** is a support for the profiles and their relationships, generally taking the form of an organized database, but includes also informal knowledge of the actors participating in the process. The memory has to be flexible and continuously updated to reflect as accurately as possible the current state of knowledge on the criminal activity [60].

The **analysis** of the memory aims at interpreting and confronting its content to all other sources of information. Profiles, links and classes are interpreted and given a meaning under the form of findings, hypotheses, recommendations or suggestions [61]. This strongly depends on the questions, such as developing hypotheses on size, structure and evolution in time and space of the phenomenon. During the analysis step, the detected data-patterns (relationships within the collected data) are interpreted beyond the basic facts to develop hypotheses on their possible causes, so-called activity-patterns (recurrences in crime activities) [65].

A crime analysis process can be explained as a transformation from data into intelligence, also called the ‘DIKI continuum’ (*data-information-knowledge-intelligence*)

[66]. In the outlined forensic intelligence process data and information are the synonyms for traces and signs which are collected, stored, analysed and interpreted to become knowledge. Finally, knowledge upon which decisions are taken to generate various types of actions is called **intelligence**. Intelligence can be used at different levels; to support specific operations or investigations (tactical and operational level) or to obtain an overview of the crime mechanisms, activity and environment (strategic level) [67].

Different areas of forensic science considering objects originating from illicit markets were benefiting from the implementation of a forensic intelligence process. Combining forensic case data with other available information led to a deeper understanding on the dynamics of the markets in question. The following section highlights only a few examples from different fields of study.

Research in the field of illicit drugs, for example, showed that the use of physical, analytical and circumstantial data in a forensic intelligence perspective could add crucial information to investigations. Classes that are formed by linking several samples can give information about the scope of an organization across time and space, about the extent and scale of the drug market itself and on the risk to consumers, due to illicit products [68]. Results of an 8-year study on the chemical profiling of illicit drugs in Western Switzerland also emphasized these advantages. Almost 6000 cocaine and 3000 heroine samples were systematically analysed and compared. The results were all stored in a database. Intelligence drawn from this database suggested, that the heroin market is more organized than the cocaine market over the studied period of time and location. Both markets seemed relatively stable, yet the heroin market was less dynamic than the cocaine market. This is an indication that heroin lasts longer on the market until it is replaced [69]. The gathered intelligence clearly provides a better understanding on the structure and functioning of the two markets.

Bächler et al. proposed a forensic intelligence model in the field of false identity documents. The process consisted of a systematic acquisition, integration and analysis of relevant information [61]. In a later study, visual features extracted from more than 300 false identity documents were codified into forensic profiles. Some of the documents were known to originate from the same sources. It was possible to assess intra-source (documents from common sources) and inter-source (documents from different sources) variations. Similarity scores were computed and compared using different metrics. The

authors were able to show a clear distinction between the intra- and inter-source similarity scores. Intra-source variations were higher and much less dispersed, indicating a limited variability of production in compliance with the extracted characteristics. On that account, it was possible to distinguish false documents based on a common or different source. Additionally, it was possible to validate the forensic profiling process and hereby the possibility to integrate the process into a forensic intelligence model [70]. The authors suggest a general application of the model for areas facing similar problems [61]. The forensic intelligence process may be broadened to counterfeiting problems, faced by industries and authorities.

A growing and very serious problem is the counterfeiting of pharmaceuticals. Tremendous benefits, the lack or even absence of legal sanctions and the complexity of the market contribute to the broadening of the phenomenon. It was realized that the structured use of analytical data could provide further intelligence insights [71]. As laid out in a study [72], profiling different seizures, using information on the chemical composition and packaging data, provided a better understanding of the production and distribution chain. 33 seizures of commonly counterfeit capsules were analysed, and the information was structured in a database. Spatiotemporal information was also added to the memory. A great number of links at different levels indicated a main network, acting on a transnational scale over a period of several years. Furthermore, the authors were able to propose a geographical order of the chemical production, the packaging, wholesalers and the final sale. In a further study [73], the authors explored the analysis of the packaging and of the chemical composition from six seizures of suspected counterfeit medicines. The purpose of this study was, in a first step, to determine whether a sample is a genuine product or a counterfeit. A secondary objective was the study of all the gathered information, from a forensic intelligence perspective. All the six studied cases were linked, either by packaging data, chemical data, or both. It was concluded, that the counterfeiters behind the analysed samples belong to the same network. Additionally, these results were confronted to available seizure information and it was possible to obtain relevant information about the modus operandi of the counterfeiters.

The continuous analysis and monitoring of counterfeited watches could also benefit from the implementation of a forensic intelligence process [59]. Counterfeit watches can

be linked if they share for example a common chemical composition or physical features. Comparable features of different objects indicate that they are repetitive effects of a common cause. The common cause is derived from the hypothesis of a common fabrication method, manufactory or at a more uncertain level the implication of a particular crime group or network [60].

2.2.2 Previous studies conducted on counterfeit watches

Counterfeiting can be subdivided into a sequence of events: **production**, **distribution** and **diffusion**. Each step is leaving traces, that can provide intelligence on the relations between them [74]. Understanding the manufacturing processes and its resulting marks is a fundamental step when dealing with watch counterfeiting [59]. It is of crucial importance to comprehend the level of intelligence that traces, and information provide.

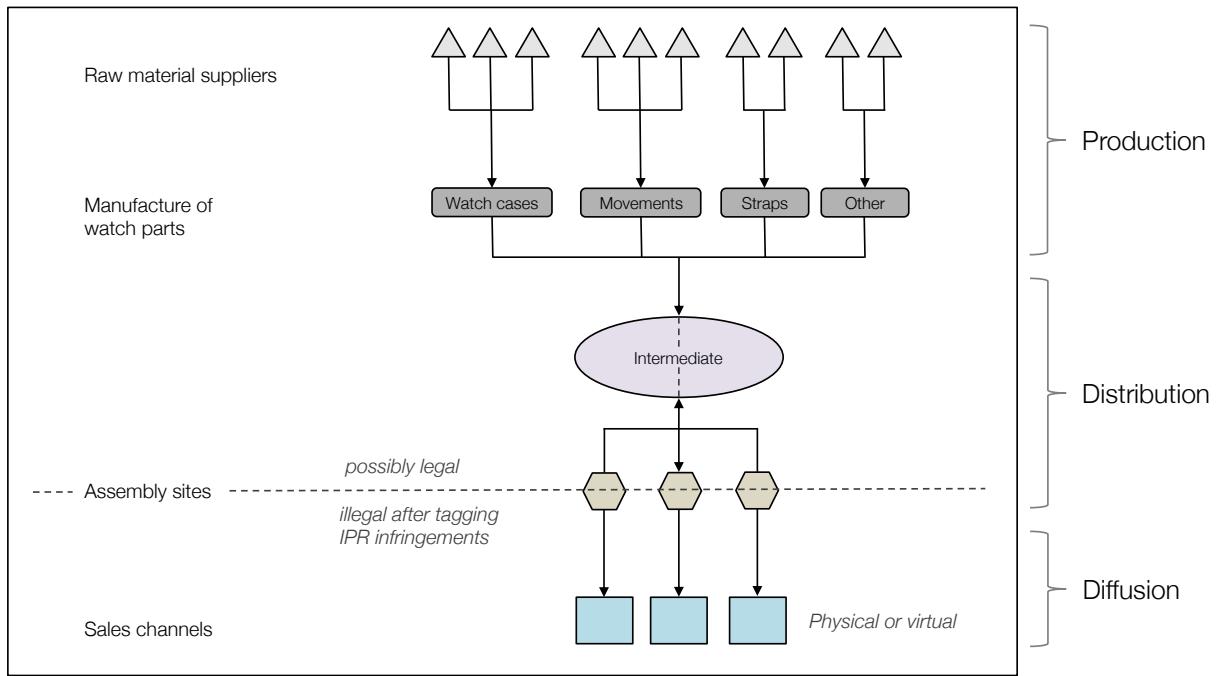


Figure 4: Example of a counterfeiting network, from Hochholdinger et al. [75]

Figure 4 shows an example of how a counterfeiting network could operate. First, there are suppliers for raw materials like metals, glass, paints and polymers. With these materials, the manufacturers produce the individual watch parts like watchcases, straps, movements and so forth. These first stages may take place in legal businesses. Individual parts can be produced for legal watch production with no intention of

counterfeiting. In a further step of this example, some intermediate buys all the different watch parts and distributes them towards multiple assembly sites, where the counterfeit watches are built together and tagged with corporate logos and brands. These assembly sites may or may not be in the same country as where the watch parts production takes place. For example, it is possible that watch parts are produced in China, and subsequently shipped to an assembly site in Turkey. The assembled and tagged watches are then sold, for example on open-air markets in attractive tourist areas. In this manner, the watch parts could cross national borders without being confiscated by custom authorities, since there is no violation of the law. Possibly, the purpose of distributing the watches towards multiple assembly sites is to not lose the whole merchandise in case of a seizure. The individual parts are then assembled, and the tagging of corporate logos and brands takes place. From this point on, intellectual property rights are violated and the line between legal and illegal business is crossed (as symbolized by the dashed straight line). In a last step, the final counterfeit watches are diffused into different markets, physical or virtual. Diffusion can take place directly from the assembly sites or by returning the final product to an intermediate.

Watch counterfeiting has rarely been subjected to scientific analysis. Nonetheless, several master theses at the ‘Ecole des Sciences Criminelles’ (ESC) of the University of Lausanne were dealing with different aspects related to watch counterfeiting. Different typologies of traces were studied, each of them providing information on a different level.

2.2.2.1 Digital traces

Previous work was conducted on the use of different digital traces. Source codes from websites selling counterfeit watches [76], online payment methods [77] and pictures used on websites, dedicated to selling counterfeit watches [78] were studied. The evaluation of these different types of information showed the possibility to draw links between different websites and to gather additional knowledge about the acting networks.

Intelligence gathered through the analysis of digital traces is located at the very end of the counterfeiting sequence, where the watches are sold. The resulting body of knowledge on the final diffusion of counterfeit watches is very useful in order to

effectively fight the sales channel ‘Internet’. Nevertheless, little information on the previous production- and distribution-chain is gained.

2.2.2.2 Package and delivery documents

The analysis of shipping documents of seized counterfeit watches by Swiss customs was also the object of a study [79]. Links were drawn based on information such as shipping location, name of the shipping company, visual resemblance of the documents, client numbers, as well as name and address of the sender.

Knowledge drawn from shipping documents was related to sales places. Furthermore, information on shipping documents might be very useful for customs on an operational level in order to target parcels likely to contain counterfeit watches.

2.2.2.3 Material traces

Further studies refer to the watches themselves and the exploitation of material traces. A research on the analysis of several watch parts (straps, watchcases, backs and dials), from a specific brand and model of a commonly counterfeit watch, revealed great variations [80]. Analysis included the use of macroscopy to identify the manufacturing method for the tagging (brand and symbols), as well as ‘Fourier Transform Infrared Spectroscopy’ (FTIR) for the analysis of varnishes and paint. The results corroborated the hypothesis raised by the FH, where individual components are produced in separated facilities and then assembled in other places. ‘X-ray fluorescence spectroscopy’ (XRF) was used in another research for the determination of the alloy composition of backs and dials [81]. Five batches of counterfeit watches that were seized in three assembling sites in China were analysed. It was found that watch parts, comprising the same production batch, were made with the same alloy. This was also the case for watches of different brands and models, which were seized in the same assembling site. It was possible to demonstrate, that chemical analysis allows revealing links between watches of different brands and models. XRF was also used to study the elemental composition of watchcases [82].

Erne et al. demonstrated in 2014 the use of chemical profiling of a restricted set of specifically perfumed plastic straps [74]. A new approach based on ‘solid phase microextraction’ in conjunction with ‘gas chromatography-mass spectrometry’ (SPME-

GC/MS) was set up. First of all, it was possible to detect volatile compounds in a reproducible manner and to define chemical profiles. They were subsequently compared with each other and grouped according to their degree of similarity. Results were integrated with the tagging information on the straps, and the spatiotemporal information on the seizures. The authors were also able to show, that straps with identical tagging had different chemical profiles. Spatiotemporal analysis revealed links between watches seized in Switzerland and other countries.

Our previous research [83] was also focused on the chemical profiling of watchstraps (plastic and artificial leather). The same SPME-GC/MS methodology was applied to a relatively large set of watch specimens ($n=97$) belonging to seizures around the world. Target compounds were selected in order to define chemical profiles and they were then compared using multivariate data analysis. This approach allowed us to identify links between watchstraps from different seizures, as well as links between watches of different brands and models. We found out, that straps produced from a same manufacturer are used for different types of counterfeit watches and are subsequently spread towards multiple locations around the world.

2.3 Toxic heavy metals in jewellery

Counterfeit goods can pose a threat to consumers' health and safety. To the present day, no published studies on potentially toxic compounds in counterfeit watches exist. Yet, several studies have been published in the field of elemental analysis of low-cost jewellery. The main object of these studies was the determination and quantification of potential hazardous and toxic elements. One of the reasons is certainly the fatal incident, leading to the death of a 4-year-old boy, from acute lead poisoning after ingestion of a metallic charm. The charm was subsequently tested, and it was found, that it contained 99.1% lead per weight [84]. It is evident that small children are particularly vulnerable, due to their lower bodyweight and the fact, that they are more likely to suck or ingest small parts.

Consumer goods from China are of particular concern. The underlining reason is the poor regulatory system, especially in terms of consumer protection legislation. Numerous products "made in China" were recalled from 'Consumer Product Safety Commission' (CPSC) in the USA, due to unreasonable health or safety risks [85]. The

lack of regulations and the potentially opportunistic use of scrap metals are the main reasons for hazardous elemental contamination in jewellery. Hence, it is important to analyse metallic parts of counterfeit watches in order to find out, if toxic elements are present. In addition, Asia is found to be the greatest production site, and China the most important source economy, for counterfeit products [11].

2.3.1 Research background for toxic compounds in counterfeit watches

As mentioned before, an initial study was conducted on watch straps [83]. A total of 97 watches were provided by the Federation of the Swiss watch industry (FH). All of them were equipped with a plastic or synthetic leather strap. The watches originated from several seizures from all over the world, notably from Greece, Paraguay, China (Guangzhou) and Dubai, and from the inventory of the FH. The specimens included counterfeit watches of several brands and models. Figure 5 provides some examples of these counterfeit watches.



Figure 5: Example of watches with plastic or synthetic leather straps used for the study

All watch straps were then analysed by solid phase microextraction in conjunction with gas chromatography-mass spectrometry (SPME-GC/MS). This analytical strategy allowed to detect volatile compounds in a reproducible manner.

Among the identified compounds, we were able to detect several types of plasticizers, antioxidants, antistatic agents and lubricants. Phthalates are used as plasticizers and

are object to an increasing number of studies, because they are partly dangerous to health. Dibutyl phthalate (DBP) is a reproductive toxicant and potentially carcinogen [86]. Diethyl phthalate (DEP) has allergen-induced asthmatic properties and is also suspected to be carcinogenic [87]. DBP and DEP were systematically found in our specimens. Another detected compound was butylated hydroxytoluene (BHT), which is used as an antioxidant and which also raises health-related concerns. It is supposed to be weakly estrogenic and carcinogenic [87].

It has to be mentioned, that the compounds were only identified and not quantified. Nevertheless, the systematic presence of these compounds found within this initial study emphasized the need of further testing. We were primarily interested in potentially harmful heavy metals, since they were already found in several types of low-cost jewellery.

2.3.2 Regulations and threshold values for heavy metals

The use of toxic heavy metal in most Western countries is regulated by specific legislation. This section considers the current legislation in terms of heavy metals, in relation with jewellery (including watches) in the European Union and Switzerland. REACH is the regulation concerning ‘Registration, Evaluation, Authorization and Restriction of Chemicals in the European Union’ [88]. Annex XVII to the REACH legislation governs restrictions for certain dangerous substances, mixtures and articles. Table 6 resumes the restrictions for heavy metals in jewellery.

State of research

| Substance | Entry | Paragraph | Legal text and threshold value |
|--------------|-------|-----------|---|
| Cadmium (Cd) | 23 | §10 | <p>Shall not be used or placed on the market, if the concentration is equal to or greater than 0.01% by weight of the metal in:</p> <ul style="list-style-type: none"> (i) metal beads and other metal components for jewellery making; (ii) metal parts of jewellery and imitation jewellery articles and hair accessories, including: <ul style="list-style-type: none"> – bracelets, necklaces and rings, – piercing jewellery, – wristwatches and wrist wear, – brooches and cufflinks. |
| Nickel (Ni) | 27 | §1b | <p>Shall not be used:</p> <ul style="list-style-type: none"> (b) in articles intended to come into direct and prolonged contact with the skin, such as: <ul style="list-style-type: none"> – earrings, – necklaces, bracelets and chains, anklets, finger rings, – wristwatch cases, watch straps and tighteners, – rivet buttons, tighteners, rivets, zippers and metal marks, when these are used in garments, <p>if the rate of nickel release from the parts of these articles coming into direct and prolonged contact with the skin is greater than 0.5 µg/cm²/week.</p> |
| Lead (Pb) | 63 | §1 | Shall not be placed on the market or used in any individual part of jewelry articles, if the concentration of lead (expressed as metal) in such a part is equal to or greater than 0.05% by weight. |

Table 6: Threshold values for Cd, Ni and Pb and legal text as specified in Annex XVII to the REACH legislation, concerning the use of cadmium, nickel and lead in jewelry

Another legal text from the European Union is the Directive 2011/65/EU, concerning the Restriction of Hazardous Substances called RoHS II [89]. Threshold values for heavy metals in accordance with Annex II to the RoHS II directive are shown in Table 7.

| Substance | Threshold value | Article and legal text |
|--------------|-----------------|--|
| Lead (Pb) | 0.1% | Article 4(1) - Member States shall ensure that EEE ¹¹ placed on the market, including cables and spare parts for its repair, its reuse, updating of its functionalities or upgrading of its capacity, does not contain the substances listed in Annex II. |
| Mercury (Hg) | 0.1% | |
| Cadmium (Cd) | 0.01% | |

Table 7: Threshold values in % by weight for Pb, Cd and Hg as specified in the Directive 2011/65/EU (RoHS II)

In Switzerland, the ‘Chemical Risk Reduction Ordinance’ (Ordonnance sur la réduction des risques liés aux produits chimiques), called ORRChim, regulates the use of certain particularly dangerous substances, preparations and articles [90]. Annex 2.18 lists the prohibitions to place on market for electrical and electronic equipment, if the concentration by mass exceeds the specified maximum concentration in the homogeneous material for the substances listed in Annex II to Directive 2011/65/EU (RoHS II). Therefore, Switzerland applies the same regulations as the European Union in terms of electrical and electronic equipment.

The content of nickel, cadmium and lead in jewellery and other metallic objects, intended to come into contact with the skin, is regulated in the ‘Ordinance on Objects Intended to Come in Contact with the Skin’ (Ordonnance sur les objets destinés à entrer en contact avec la peau) [91]. Nickel is limited to 0.5 µg/cm²/week, specifically mentioned for watchcases, straps and clasps (article 2). Cadmium is limited to 0.01% (article 2a), and lead to 0.05% by weight (article 2b). This regulation also specifically applies to wristwatches.

2.3.3 Lead

Lead (Pb) is used over a broad range of applications. Jewellery parts like clasps, charms and chains can also contain lead. Lead is neurotoxic and harmful to most human organs and systems [92].

Several studies showed the presence of lead in low-cost jewellery. Maas et al. analysed a total of 311 pieces of metallic jewellery for lead content, using flame atomic absorption spectroscopy (FAAS). The jewellery samples were purchased throughout California, and some of them were clearly intended for children. An overall mean of 30.6% lead

¹¹ The acronym EEE stands for electrical or electronic equipment and includes therefore quartz watches.

was detected and 123 samples were found to contain more than 50% lead by weight. The authors also pointed out the importance to pinpoint sources of lead exposures from consumer goods [93].

In 2007, Weidenhammer and Clement examined 139 samples of low-cost jewellery. The vast majority of the samples were imported from China, intended for children and purchased for less than USD 10. All metal samples were analysed for lead content, using FAAS. Results showed extremely high lead content. A significant percentage of samples (42.6%) contained more than 80% lead by weight [94]. The authors also raised the question, whether electronic waste and battery lead might be the source materials for lead found in inexpensive jewellery. Electronic waste, or “E-waste”, is a continuously growing problem. An enormous amount of “E-waste” is shipped from the USA, along with other rich economies, to the developing countries of East Asia, particularly China [95]. Lead/tin solders are widely used in electronic goods and recovered from “E-waste”. However, there is no indication for the use of recovered solders. Proportions found in lead/tin solders from China and India are, typically within 36.2-38.5% lead and 46.3-49.8% tin [96]. Weidenhamer and Clement found, that the overall proportion of lead in the analysed samples deviated from the expected values for the solders, with a mean of 70.1% lead and 24.6% tin. This either means, that recovered solders are not the origin of lead in jewellery, or additional lead from other sources is added [94].

Battery lead is believed to be another source. Most of the lead is used in the automotive sector, as chemical energy storage, in form of lead accumulators [97]. Hence, the objective of a further study was to establish, whether or not battery lead is used to manufacture inexpensive jewellery. Automotive battery lead basically contains 1.5-4.5% antimony (Sb) by weight. 39 highly leaded (> 90%) jewellery samples were analysed. The antimony concentration ranged between 0.3% and 6.2% by weight, with a mean of 3%. Thus, the chemical composition resembles the lead-antimony alloys in accumulators, suggesting that parts of battery lead might be reused for jewellery manufacture [98].

2.3.4 Cadmium

Cadmium is primarily used in nickel/cadmium (Ni/Cd) cells [99]. However, cadmium can also be used as surface paint, plating or even as a base material in jewellery.

Particularly strong attention is drawn on the regulation of cadmium in children's products. Restrictions on the use of cadmium in the European Union caused a price reduction due to dropping demand of Ni/Cd cells. Product manufacturers might choose cadmium as a substitute [92]. Cadmium is a known carcinogen and can cause kidney disease [100].

High levels of cadmium were already found in low-cost jewellery. A study assessed the total metal content of 21 specimens of silver jewellery from India, seized from the noble control department of Zurich airport. These products were seized due to high levels of cadmium detected by portable XRF instrument. The metal content was analysed by AAS and ICP-MS. The concentration of cadmium was the second highest, after silver, ranging from 1.4 to 43.9% [99]. In another study conducted in 2011, several jewellery components were selected, based on previous XRF screening, showing high Cd concentrations. Package labelling showed, that nearly all the probed jewellery was made in Asia, mostly in China. Total Cd concentrations were analysed using AAS, determining an overall average concentration of 39.27% [101].

2.3.5 Miscellaneous

Guney and Zagury investigated the contamination of ten harmful elements in toys and children's jewellery. Analysed elements were arsenic (As), barium (Ba), cadmium (Cd), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), lead (Pb), antimony (Sb) and selenium (Se). These elements are regulated in the 'EU regulation on the safety of toys'¹² [102]. In total, 24 metallic toys and jewellery were tested, using ICP-OES. It was found, that 20 samples exceeded the EU migration limits, for at least one element, on the total metal concentration. Violations of limits were mostly found for Cd, Cu, Ni and Pb. In some samples, the limits were exceeded by a factor of more than 1000.

2.4 Knowledge gained through previous research

Watch counterfeiting involves a series of interconnected steps, each of them leaving traces and information. The relationships between these steps are currently not very

¹² Limits are given in terms of migration [$\text{mg}\cdot\text{kg}^{-1}$]. The authors assume, if the total metal concentration exceeds the limit for a metal, further migration testing is needed.

well understood. Previous studies on counterfeit watches emphasized the potential to gather information from several sources. Digital traces and shipping documents revealed intelligence on the acting networks that sell counterfeit watches. Approaches based on material traces delivered additional knowledge on the production of watch parts and their distribution towards assembly sites.

Previous studies corroborated the following hypotheses:

- Individual parts are produced in separate facilities and mounted in different assembly sites.
- Chemically linked watch parts (dials, backs and straps) are used for the production of counterfeit watches of different brands and models.
- Watchcase production takes place on a larger scale than the subsequent tagging and assembling of the watches.

These preliminary studies emphasized the need and importance of contextualizing all available datasets in order to study the relation between production and distribution, and therefore producing useful intelligence on the market of counterfeit watches.

Reviewed literature on inexpensive metallic jewellery indicated large quantities of potentially toxic heavy metals. These studies refer only to low-cost jewellery and the results cannot be generalized to (low-quality) counterfeit watches. Chemical analysis of watchstraps revealed the presence of phthalates and other potentially toxic compounds. Whether toxic heavy metals are present in other watch parts, for example watchcases, has not been studied yet.

3 Research goals

3.1 Definition of the research goals

The goal of this PhD thesis was to assess the contribution of chemical and physical analysis of watches in order to gather actionable knowledge on the market of counterfeit watches. The approach aims to apprehend activities, based on the trafficking of counterfeit watches, through contextualization of information and traces. Understanding the relationships between the production- and distribution steps is essential to develop strategies to counteract. New insights into the structure and organization of the illicit market of counterfeit watches could support decision-making, in terms of priorities of investigations, and possible preventive measures to be implemented.

The studied information will be of three different dimensions:

- **Chemical profiling** using the results of elemental analysis of watchcases
- **Physical profiling** based on the codification of physical characteristics of all imprints present on the counterfeit watches provided by the Federation of the Swiss Watch Industry (FH)
- **Spatiotemporal information on seizures**, gathered from the FH reports on seized watches

A further and complementary objective of this research is to appraise the **potential risk** to public health due to toxic heavy metals in watchcases and its relevance as an anticounterfeiting approach.

3.2 Sampling and selection of watch parts

The first specimen set consists of 35 watches of the same brand, including seven models. The brand (Rolex) was chosen because it is one of the most commonly counterfeited. Considering watches of only one brand, seized during a relatively short period of time (seven months), increases the chances of finding links (on a chemical and physical level) between them. It is more likely that the same raw materials and the same tools for tagging the watches were used. The replica manufacturer typically buys large quantities

of individual parts needed to assemble a watch. It is assumed that the same pieces are mounted until the stocks are exhausted [103].

Only watchcases were chosen as the object of study. The potential risk to consumers due to the presence of toxic elements is higher in watch parts, which are in direct and prolonged contact with the skin. Watchstraps were not taken into account, because they are made of different materials such as leather, artificial leather, plastic and metal. Thus, it would be difficult to establish a single method for chemical analysis. Watchcases on the other hand, are almost exclusively made of metal.

The manufacturing of metallic pieces became easier through the use of computer automation for machine tools, called CNC¹³. CNC machines are used to cut out cases and other watch parts from solid pieces of metal [103]. It is possible to reconstruct exact copies of the shapes of watch parts. However, we assume that only a finite number of production sites, intending to manufacture copies of watch parts, have access to such technology. It is more likely that watchcases originating from a same production batch have the same chemical composition. This increases the chances of finding links between watchcase producers. Hence, more information on acting networks can be gathered.

The second specimen set is composed of watches of various brands and of different degrees of quality levels. These watches were chosen to determine the presence of potentially toxic heavy metals. No further exploitation was considered for this particular specimen set.

For further information on the specimen sets of counterfeit watches used for this research see section 4.2 as well as annex 11.1 and 11.2.

3.3 Definition of terminology

Throughout the previous chapter important notions such as ‘trace’, ‘profile’, ‘link’, ‘class’ and ‘group’ were introduced. However, it is important to clearly define what shall be understood by these notions in the specific context of this research.

The studied traces are of different nature, namely chemical and physical traces.

¹³ This abbreviation stands for ‘Computer Numerical Control’

- **Chemical trace:** The trace we were looking for is the elemental composition of watchcases. Even though, strictly speaking, the elemental composition reflects the alloy production itself, it can indirectly be seen as a remnant of the production of the watchcase.
- **Physical trace:** During the physical analysis of counterfeit watches at the FH, all imprints present on the different parts of the counterfeit watches were examined independently by the specialists at the FH. An imprint is a physical mark that is left on the watch, that either shows a text or a symbol, representing a brand, a model, a geographical, technical or quality-related indication. Imprints are the physical traces left from the tools that were used to produce them.

As further detailed in this chapter, chemical and physical features are extracted in order to draw profiles that must be relevant to the context.

- **Chemical profile:** The chemical profile is constructed by selecting relevant attributes within the elemental composition.
- **Physical profile:** The physical profile is a numerical code identifying each imprint. Imprints with the exact same characteristics such as position, manufacturing process, category, content of the inscription and measurement values of width and length are translated by the same code.
- **Chemical link:** The degree of relationship between profiles is measured and expressed as a score. A chemical link between profiles is defined on the basis of a threshold value. Relationships between more than two profiles are defined as a **group** or a **class**¹⁴. These terms are used synonymously in the context of this research.
- **Physical link:** Two watches that contain the same codes are linked via this code. Hence, two watches can be linked by several codes on different parts.

The **grouping** of the physical profiles is a more difficult task since several tools can be used to tag imprints onto different watch parts. It is necessary to search for relevant configurations between specific types of imprints on specific watch

¹⁴ In chemometrics the notion of ‘classification’ is, strictly speaking, used for supervised learning models where the algorithm is trained on a labeled dataset. This was not the case for the available data.

parts.

These notions being introduced, the methodological approach as well as research hypotheses are further detailed.

3.4 Chemical information: elemental analysis

Watch counterfeiting is a complex, multi-step phenomenon and the structure of the market is still not very well known. Difficulties also arise due to the distribution towards multiple assembly sites and the diversification of the sales channels. The elemental composition of watchcases is independent from these steps and could fill the information gap on the starting point, which is the production of the watch parts.

As Margot [54] stated: the shorter the time elapsed or the less traces are fragile, the more accurate is the story that traces tell. The problem of particular fragile traces does not appear to be a major issue for the elemental analysis of metals, yet the time elapsed since the production of the watchcase is. Elemental composition could be different if the production batch changes. This would inevitably lead to a differentiation of the watchcases. It is therefore more meaningful to consider watches that were seized during a short period of time. It is more likely that watches seized during a short lapse of time are closer to their respective production period, even though the time of production is not known.

3.4.1 Data acquisition: development of an analytical strategy

A suitable analytical strategy had to be elaborated in the first place. This raised several questions and challenges.

Down to the present day, no published reference in the field of chemical analysis of watchcases (or any other metallic parts from watches) was found. Therefore, literature review was mainly based on the analysis of metallic jewellery.

A further aspect of interest was the use of substandard materials in counterfeit products that potentially present a threat to public health. Studies showed that harmful heavy metals, like lead and cadmium, can be found at high concentrations in low-cost jewellery (see section 2.3). The hypothesis can be proposed that such elemental contamination can also be found in counterfeit watches. Furthermore, potentially harmful compounds were already detected in watchstraps [83].

In order to collect information on the production of the watches, elemental analysis of the watchcases was adopted as a method of choice. Several analytical methods exist in the field of elemental analysis. For solid samples, the most commonly used analytical techniques are X-ray fluorescence spectrometry (XRF) and atomic spectroscopic techniques.

XRF is a multi-elemental analysis with a broad range of applications. It has already been used in the field of jewellery analysis [99, 101, 104-107]. XRF has several beneficial characteristics, especially in terms of a screening technique: it is a multi-elemental technique, easy to use, non-invasive and non-destructive. The non-destructive character of XRF is particularly interesting for the analysis of counterfeit watches, because in some cases, it might be necessary to preserve the integrity of the watch. Furthermore, field portable instruments also exist.

Several atomic spectroscopic techniques exist. Some of the most commonly used are: flame atomic absorption spectroscopy (FAAS), graphite furnace atomic absorption spectroscopy (GFAAS), inductively coupled plasma optical emission spectroscopy (ICP-OES), inductively coupled plasma atomic emission spectroscopy (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS)[108].

ICP-MS has already been in use for analysing jewellery [99, 104] and for the analysis of trace metals in steel [109]. ICP-MS is also one of the suggested techniques in the CPSC Standard Operating Procedure for the determination of lead and cadmium in children's metallic products [110, 111]. ICP-MS offers several advantages; it is a quantitative, multi-elemental technique with a sensitivity allowing trace and even ultra-trace analysis. Clearly, the destruction of the sample through acid digestion is a disadvantage of ICP-MS compared to non-destructive techniques, such as XRF analysis.

Two types of elemental analysis were chosen in order to study the alloy composition of watchcases. The first one is XRF, as a screening method. The second method of choice is ICP-MS after acid digestion. Figure 6 shows the sequence for the watchcase analysis.

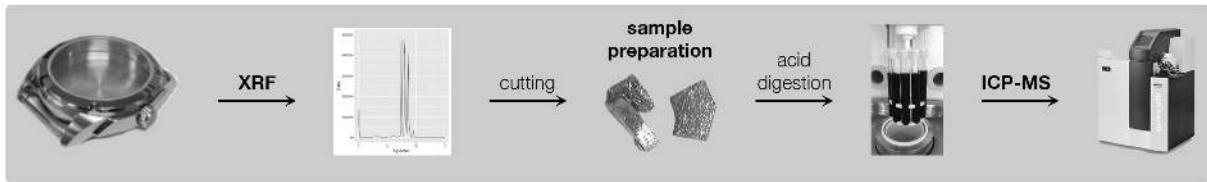


Figure 6: Sequence for the elemental analysis of watchcases

The first hypothesis of this research was:

-
- H1:** The analytical approach enables to obtain reliable data on the chemical composition of watchcases.
-

In order to test this hypothesis, the specimen set of watchcases had to be relatively small. There was no point in developing and testing a new method on a very large specimen set. This also justifies the first specimen set, consisting of 35 watches.

XRF results were evaluated as an analytical technique on its own, as well as its utility as a screening technique prior to ICP-MS. Furthermore, the agreement between the two methods of measurements was determined.

3.4.2 Chemical profiling through multivariate data treatment

The aim of the chemical profiling is to link watchcases from a common source and to discriminate them from watchcases from different sources.

Target compounds need to be selected from the analytical results, and the chemical profiles have to be defined. It is possible to measure the similarity (e.g. distance or correlation measurements) between profiles. The initial data may need basic pre-processing, like centering, scaling or normalization to obtain a better symmetry. Data pre-processing should be part of the overall approach. Multiple pre-processing techniques exist, and it is important to understand their impact [112].

A binary classification model can be created by measuring the similarity of reference data, namely profiles that are known to originate from common, respectively different sources. In this regard, intravariability and intervariability distributions can be determined and serve as a basis for the determination of a threshold value of the similarity score to decide whether two specimens are linked or not based on their chemical composition. In practice, there is usually some degree of overlap between

intra- and intervariability distributions. The overlapping zone is an area of uncertainty where the risk of false attributions can be quantified.

Threshold values strongly depend upon the context [113]. From a judicial perspective, the strategy is to restrict as much as possible the false positive (FP) rate and at the same time the risk of producing erroneously incriminating evidence. Considering an intelligence-based perspective, the goal is to minimize the false negative (FN) rate [62]. Sensitivity (true positive rate) and specificity (true negative rate) can be studied at a given threshold. Another performance criterion of a binary classification model is the receiver operating characteristic (ROC) curve. The ROC curve requires plotting the true positive rate against the false positive rate as the threshold changes and the Area Under the Curve (AUC) is considered as a measurement of the accuracy of predictive distribution models.

Large amount of data, containing several elements, has to be handled. Multivariate analysis aims to explore relationships and patterns between multiple variables, measured for each specimen and to determine the relationships in form of a mathematical model [114]. For multivariate datasets the challenge is often to reduce the data dimensionality to gain comprehensive information [115].

In exploratory data analysis (EDA) the aim is to visualize the relationships between specimens and between variables that are not necessarily assigned to predefined groups and hence a preliminary stage prior to supervised modelling. EDA can help to answer questions such as whether there is some kind of grouping in the data or whether samples from a similar source are related [112]. Principal component analysis and cluster analysis are initial and recommended steps for a better insight in the structure of the data [114].

Principal Component Analysis (PCA) reduces the initial number of dimensions, making it possible to represent them graphically. A new coordinate system is computed. In PCA the original variables are transformed into new, non-correlated variables that are linear combinations of the original ones. These new variables are called the principal components (PC_1, PC_2, \dots, PC_n). PCA is equivalent to an axis rotation, where PC_1 directs towards the greatest variation, PC_2 towards the next greatest variation, and so forth, until including all the information with PC_n . The information content conveyed by the raw dataset can be represented in 2 or 3 dimensions, instead of the initial n

dimensions, losing only a small amount of information [115]. Correlated variables are an indication of redundancy in the data, hence PCA can be used to identify such variables and to remove them.

Cluster analysis is based on grouping specimens into clusters, according to similarity measures. Specimens that display similar characteristics in the variable space are grouped in the same class. The distance between two points in an n-dimensional space is commonly computed using the Euclidian distance, knowing that other distance or correlation measures exist. There are also several clustering methods [115]. A commonly used method is ‘hierarchical cluster analysis’ (HCA). In this approach, specimens are portioned into k clusters ($k = 1, \dots, n$). The grouping is visualized in a hierarchical dendrogram, where the x- or y-axis displays the distance between two points. Specimens from a cluster of a lower level are a subset of those from higher levels in the hierarchy [114]. Therefore, the distance, at which the grouping is stopped, determines the number of final clusters. A commonly used method for HCA is Ward’s minimum variance criterion. This method searches the pair of clusters leading to minimum increase of the total within-cluster variance when they are merged [114].

The second hypothesis of this research is:

H2: The analytical results allow for the grouping of specimens into classes according to their chemical resemblance.

Different types of profiles were taken into account, testing different sets of variables.

3.4.3 Knowledge gathered through the study and interpretation of chemical links

The goal of the chemical profiling part was to group the specimens according to the similarity of their elemental composition and hence provide new knowledge on the production of these watches. The underlying hypothesis claims that a chemical group or class of specimens is representative of a common source of alloy production.

However, it has to be kept in mind, that chemical links between two watchcases are based on the level of the alloy production itself, and not strictly on the watchcase

production. The same alloy manufacturer could possibly supply several different watchcase producers.

A great variety of alloys could be an indication for numerous different producers, or an opportunistic use of the source material. On the other hand, a small variety could indicate, that there are only a few producers, or that the alloy compositions are very close.

The third hypothesis of this research is:

-
- H3:** Knowledge drawn from chemical profiling provides new insights on the production level of watchcases.
-

3.4.4 Possible presence of toxic heavy metals

Analysis was also focused on the detection of potentially hazardous heavy metals. Especially for this part the ultra-trace capabilities of ICP-MS or the portable character of the XRF machine may be beneficial. Nonetheless, the research question was not to quantify hazardous elements if they were present, but to generally evaluate a potential risk. If hazardous elements were to be found, the fight against watch counterfeiting could also be seen from a public health and safety angle. According to reviewed literature on jewellery analysis, several toxic heavy metals are regularly found in low-cost jewellery.

The fourth hypothesis of this research is:

-
- H4:** Watchcases contain potentially toxic heavy metals.
-

Special emphasis was laid on heavy metals regulated by the Swiss law, namely lead (Pb), cadmium (Cd) and nickel (Ni).

We expected to find more elemental contamination in poor quality watches. The first specimen set only contains watches of relatively good quality, large amounts of potentially toxic heavy metals were therefore not expected. Another specimen set was specially chosen for this part, including also mid-range and poor-quality copies to test the underlying hypothesis.

3.5 Physical and spatiotemporal information: Codification of imprints and seizure information

3.5.1 Methodology applied by the FH

This research was conducted in close collaboration with the Legal and Anticounterfeiting Department of the Federation of the Swiss Watch Industry (FH). The FH provided all counterfeit watches used for this research with the corresponding expertise reports and allowed access to all relevant seizure information in their database.

Specimen set 1 was examined by specialists at the FH and a total of 19 expertise reports have been issued for the 35 watches, since some watches were being seized together. These reports contain information about the inventory and the seizure, the general examination, identification of the clockwork, identification of imprints and a list of connections that were generated within the database with formerly analysed watches. Links between watches are therefore based on physical relationships. The working scheme for this procedure is illustrated in Figure 7.

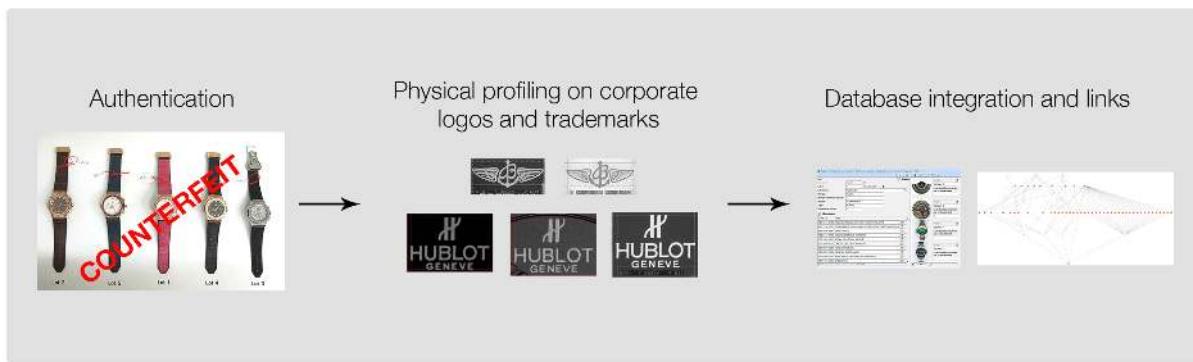


Figure 7: Working scheme for the analysis of watches at the Anticounterfeiting Department at the FH

It is obviously important to identify and list the connections that a particular counterfeit watch shares with formerly analysed watches. Nevertheless, the information content that links between imprints convey is currently underexploited by the FH.

3.5.2 Knowledge gathered through the study and interpretation of the physical links

All imprints present on the different parts of the counterfeit watches were examined independently by the specialists at the FH: strap, clasps, watchcase, dial and the crown. The different watch components on which imprints can be found are shown in Figure 8.

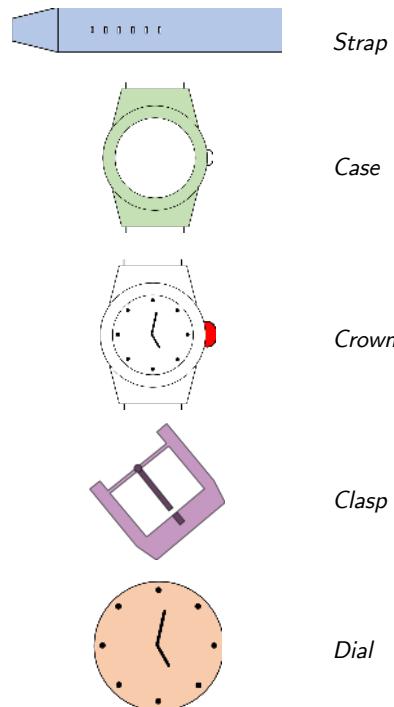


Figure 8: Examined watch components on which the imprints can be found

The imprints that can be found on counterfeit watches can be divided into several categories; either representing brands, logos, imprints containing qualitative or technical information or an indication of a source. Examples of each category can be found in Figure 9.

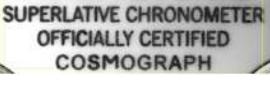
| <i>Brand</i> | <i>Logo</i> | <i>Qualitative</i> | <i>Technical</i> | <i>Source indication</i> |
|--|---|---|--|---|
|  HUBLOT GENEVE |  |  |  |  |

Figure 9: Examples of different types of imprints that can be found on counterfeit watches

Information on these imprints are translated into numerical codes, that are introduced into their database. These codes contain different types of information, translated by a number:

- Position of the imprint (case, dial, crown or strap)
- Manufacturing process (e.g. struck, engraved)
- Category (e.g. symbol, brand)
- Content of the inscription
- Measurement of length and width of the imprint

These numbers, concatenated into a single code in the form of XXX.XXXX.XXXX, identify each imprint. Subsequently, the codes are used to search the database and make comparisons to detect the links between identical imprints (e.g. identical codes). A link suggests that two imprints were produced by the same source, i.e. the same manufacturing tool.

Various types of information can be gathered in addition to the fact that a particular watch is linked to another one through a particular imprint.

The fifth research hypothesis is:

-
- H5:** A thorough study of all imprints found on the specimens, as well as their relationships with previously analysed watches creates new information and insights. This allows for a deeper comprehension on the use and scope of the tagging tools and on the structures of the underlying assembly sites.
-

3.6 Forensic intelligence through contextualization

The suggested methodology aims to obtain information at two levels. First of all, the elemental composition of the watchcase, which is related to its production. Secondly, the physical profiling of logos and trademarks provide information on the distribution towards different assembly sites. The FH reports on the examined counterfeit watches contain spatiotemporal information, including place and date of seizure, and origin and ways of transit, if known. Place of seizure, origin and ways of transit allow further investigation concerning the geographic distribution of the counterfeit watches. Dates

of seizure can be subjected to a temporal analysis. Hence, every source of information should result in additional knowledge, but on a different level. The central part of this research will be the contextualization of all the gathered information.

The mere elemental composition already provides information on the production process. Chemical classes can give information on the magnitude of watchcase producers, whereas physical classes shed light on the spread of assembly sites.

Figure 10 shows at which level the different sources of information could add new knowledge.

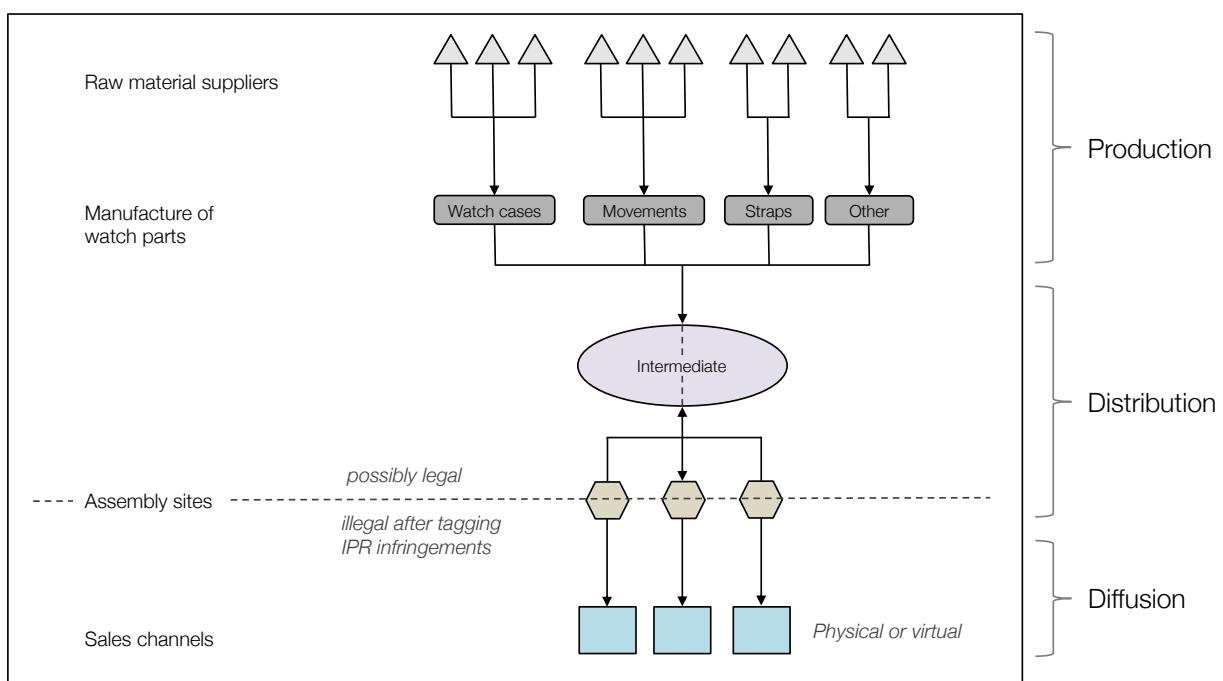


Figure 10: Counterfeiting network and added information at each level

The study of the physical profiling reveals links and information on a different level than the watchcase analysis. Since tagging does supposedly not take place in the production facilities of the individual watch parts, a prior distribution towards the assembly sites occurs. Links between physical characteristics inform, if the tagging was performed by a same tool and probably in the same assembly site. Analysing several watches from the same brand and model, seized during a relatively short time, can help to estimate the scope of assembly sites tagging the same kind of watches.

Combining chemical and physical information leads to the sixth research hypothesis:

-
- H6:** The comparison between the chemical and the physical profiles provides valuable intelligence on the distribution of counterfeit watches towards the assembly sites, resulting in a deeper comprehension of the relations between these two steps.
-

The limiting factor is, however, the specimen set size. It will not be large enough to allow generalized market conclusions. Nonetheless, the specimen set is very interesting since it is based on a relevant selection of counterfeit watches of a single brand seized during a defined time window. The specimen set constitutes the prototype to study the different processes and the potential to gather intelligence.

4 Experimental section

4.1 Overview

Experimental details for all work carried out over the course of this thesis can be found in this chapter, this includes relevant details in relation to specimen selection, analysis and validation.

The two following specimen sets were used:

- **Specimen set 1:** consisting of 35 counterfeit Rolex watches
- **Specimen set 2:** consisting of 35 counterfeit watches of various brands to test for compliance with the current legislation in Switzerland regarding potentially toxic heavy metals

For the two sets, the following experimental analyses were conducted:

- Bench-top XRF screening analysis¹⁵ [82] of **Specimen set 1**
- ICP-MS analysis of **Specimen set 1**
 - o Analysis of major elements
 - o Analysis of minor elements
- Portable XRF analysis of **Specimen set 1** and **Specimen set 2**

4.2 Specimen sets

4.2.1 Specimen set 1

Specimen set 1 consisted of 35 counterfeit watches of the brand ROLEX SA and included seven different models. The Rolex brand was selected because these watches are some of the most commonly counterfeited in the luxury watch segment as well as being one of the most counterfeited brands in general. The watches were all seized by customs authorities in Switzerland in 2015 and their respective model and their ID-numbers are listed in Table 8. A detailed description, as well as photographic documentation can be found in the annex (11.1 and 11.2). The watches investigated in

¹⁵ Conducted as part of a Master thesis in 2016 by Sandra Langel

Experimental section

this thesis had been previously disassembled by FH specialists and only their cases were retained for study.

| Number | Number SAC | Brand | Model |
|--------|------------|-------|-----------------------|
| M01 | 15916 | Rolex | Deepsea (Sea-Dweller) |
| M02 | 16008-1 | Rolex | Submariner Date |
| M03 | 16008-2 | Rolex | Cosmograph Daytona |
| M04 | 16018-1 | Rolex | Cosmograph Daytona |
| M05 | 16018-2 | Rolex | Submariner Date |
| M06 | 16029-1 | Rolex | Submariner Date |
| M07 | 16029-2 | Rolex | Submariner Date |
| M08 | 16030 | Rolex | Submariner Date |
| M09 | 16093-1 | Rolex | Cosmograph Daytona |
| M10 | 16093-2 | Rolex | Cosmograph Daytona |
| M11 | 16094-1 | Rolex | Cosmograph Daytona |
| M12 | 16094-2 | Rolex | Cosmograph Daytona |
| M13 | 16095-2 | Rolex | Cosmograph Daytona |
| M14 | 16096-1 | Rolex | Cosmograph Daytona |
| M15 | 16096-2 | Rolex | Cosmograph Daytona |
| M16 | 16102-1 | Rolex | Datejust |
| M17 | 16102-2 | Rolex | Datejust |
| M18 | 16103-1 | Rolex | Submariner Date |
| M19 | 16103-2 | Rolex | Submariner Date |
| M20 | 16105-2 | Rolex | Datejust |
| M21 | 16106-2 | Rolex | Cosmograph Daytona |
| M22 | 16107 | Rolex | Air-King |
| M23 | 16104-1 | Rolex | Day-Date |
| M24 | 16104-2 | Rolex | Day-Date |
| M25 | 16095-1 | Rolex | Cosmograph Daytona |
| M26 | 16097-1 | Rolex | Cosmograph Daytona |
| M27 | 16097-2 | Rolex | Cosmograph Daytona |
| M28 | 16098-1 | Rolex | Day-Date |
| M29 | 16098-2 | Rolex | Day-Date |
| M30 | 16099-1 | Rolex | Day-Date |
| M31 | 16099-2 | Rolex | Day-Date |
| M32 | 16100-1 | Rolex | Day-Date |
| M33 | 16100-2 | Rolex | Cosmograph Daytona |
| M34 | 16105-1 | Rolex | Datejust |
| M35 | 16106-1 | Rolex | Yacht _ Master II |

Table 8: Listing of all watches from the main Rolex specimen set, their respective model and allocated ID-numbers for this study (MXX) and for the physical expertise at FH (number SAC)

4.2.2 Specimen set 2

For the portable XRF analysis an additional specimen set, called *specimen set 2*, that included 35 counterfeit watches of various brands was selected. For this specimen set, an attempt was made to cover the broadest range of counterfeit watches. Based on visual inspection by the experts at the FH, the watches were divided into three levels of quality: high (n=10), average (n=13) and low (n=12) quality counterfeits (Table 9). As a first step, all the watches were disassembled and only the watchcases, straps (with clasps) and watch backs were retained for further analysis. *Specimen set 2* contained counterfeit watches of 14 brands that were seized at different times. The watches in this set were selected for elemental composition analysis in order detect whether or not potentially toxic elements were present on the surface of counterfeit watches. No further analyses were undertaken for this specimen set.

Experimental section

| Quality | Number | Number SAC | Brand | Model |
|---------|--------|--------------|-----------------|--------------------|
| High | M01 | 15405 | Hublot | Classic Fusion |
| | M02 | 15412-1 | Hublot | Big Bang |
| | M03 | 15404-2 | Hublot | Classic |
| | M04 | 15417-1 | Patek Philippe | 5002 |
| | M05 | 15417-2 | Patek Philippe | - |
| | M06 | 15433-2 | Tag Heuer | Carrera |
| | M07 | 15448-1 | Audemars Piguet | Royal Oak |
| | M08 | 15448-2 | Audemars Piguet | Royal Oak |
| | M09 | 15432-1 | Tag Heuer | Diamond Star |
| | M10 | 15432-2 | Tag Heuer | Diamond Star |
| Average | M11 | 14978 | Breitling | Bentley |
| | M12 | 14424 | Breitling | Bentley |
| | M13 | FH inventory | Hublot | Classic fusion |
| | M14 | 14586-2 | Hublot | Classic fusion |
| | M15 | 14587 | Hublot | King Power |
| | M16 | FH inventory | Hublot | Big Bang |
| | M17 | 14604 | Ulysse Nardin | Marine |
| | M18 | FH inventory | Ulysse Nardin | Marine |
| | M19 | 14585 | Patek Philippe | - |
| | M20 | 14775 | Rolex | Datejust II |
| | M21 | 14600 | Tag Heuer | Mikrotourbillon |
| | M22 | 14369 | Franck Müller | Elegance |
| | M23 | 14980 | Franck Müller | Crazy Color Dreams |
| Low | M24 | 15056-1 | Tissot | PRC 200 |
| | M25 | 15059-2 | Tag Heuer | - |
| | M26 | 15072-2 | Chopard | - |
| | M27 | 15063-2 | Hermès | Arceau |
| | M28 | 9520 | Longines | Grande Classique |
| | M29 | FH inventory | Audemars Piguet | - |
| | M30 | 6760 | Omega | Constellation |
| | M31 | FH inventory | Rolex | Datejust |
| | M32 | 4984 | Rolex | Submariner |
| | M33 | 15073-2 | Patek Philippe | Sky Moon |
| | M34 | FH inventory | Gucci | - |
| | M35 | 6858 | Gucci | - |

Table 9: Listing of all watches of specimen set 2, their quality, respective model and allocated ID-numbers for this study (MXX) and for the physical expertise at FH (number SAC)

4.3 Bench-top XRF analysis on *specimen set 1*

The elemental compositions of the watchcases were obtained by bench-top X-ray fluorescence analysis as a part of a master thesis undertaken in 2016 [82], and the measurements were carried out prior to ICP-MS analysis. Non-destructive semi-quantitative analysis was conducted using a XGT-7000 micro XRF-analyser (Horiba, Kyoto, Japan) with a rhodium X-ray tube set at 30kV. For the measurements, full vacuum conditions were used, and the spot size was set at 100 µm. The specimens were analysed without any prior preparation and four measurements were performed for each watchcase. No certified reference materials were used during this study; therefore, only semi-quantitative results were obtained.

These preliminary results were solely used for alloy identification, which is an important step prior to ICP-MS analysis, as explained below. All the watches were made of austenitic stainless steel, except for specimen *M01*. Austenitic steel is designated as alloys with Fe, Cr and Ni as the major constituents [116].

4.4 ICP-MS analysis of *specimen set 1*

4.4.1 Analytical procedure

4.4.1.1 Reagents

- Nitric acid (HNO_3 , > 69.0%, TraceSelect, Fluka Analytical)
- Hydrochloric acid (HCl , > 37%, TraceSelect, Fluka Analytical)
- High purity argon gas (purity > 99.999%, ALPHAGAZ)
- Certified single element standards:
 - aluminium (Al), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd), tin (Sn), lead (Pb)
 $1000\mu\text{g}\cdot\text{mL}^{-1}$, CentriPUR, Merck
 - phosphorus (P), silicon (Si), titanium (Ti), vanadium (V), molybdenum (Mo), tungsten (W)
 $1000\mu\text{g}\cdot\text{mL}^{-1}$, TraceCert, Sigma-Aldrich

- uranium (U)
1000 $\mu\text{g}\cdot\text{mL}^{-1}$, Inorganic Ventures, Inc.
- Internal standards
 - scandium (Sc), germanium (Ge), yttrium (Y), rhodium (Rh), indium (In), terbium (Tb), bismuth (Bi)
1000 $\mu\text{g}\cdot\text{mL}^{-1}$, CentriPUR, Merck
- Ultrapure water (18.2 M Ω cm) prepared with an ELGA PURELAB instrument

4.4.1.2 Sample preparation

The preparation of our specimens for ICP-MS was generally challenging. For analysis, the sample has to be in a liquid state¹⁶ and a simple method for this is to dissolve the sample in a strong acid. It is generally recommended to start digesting with diluted (5-10%) nitric acid (HNO_3). If the sample does not completely dissolve, concentrated nitric acid can be added. Alternatively, a mixture of nitric acid and hydrochloric acid (HCl) can be used [117]. Digestion can take place in an open vessel, however, dissolving a sample in a closed vessel by microwave digestion is more advantageous. This allows for reproducible digestion conditions through the automated control of the temperature and pressure as well as reduced airborne dust contamination; in addition, toxic fumes are eliminated, and less digestion solution is needed.

Since it is not possible to dissolve the entire watchcase, small cuts of approximately 1g were obtained by electrical discharge machining. This step was conducted by the precision mechanics workshop at ‘Ecole Polytechnique Fédérale de Lausanne’ (EPFL). Watchcases are frequently plated, cheap counterfeits for example, are often plated with a very thin layer of gold. A certain watch model might be available in two different versions, such as gold and silver. However, in the case of different versions for counterfeit watches, the underlying watchcase might consist of the same alloy composition, but with different plating for the top layer. Therefore, it is important to include a cleaning step in order to get rid of the plating. It is estimated that a 20% weight reduction is sufficient in order to eliminate the entire surface plating, as well as

¹⁶ Solid samples can be analysed by laser ablation ICP-MS (LA-ICP-MS)

other surface contaminants. This cleaning step takes place in an open vessel and the entire sample is subsequently dissolved using microwave digestion.

A series of tests using nitric acid (HNO_3) and hydrochloric acid (HCl) were performed in order to establish a cleaning and digestion procedure. The sample cuts were precleaned in open Falcon tubes (15ml) containing a 3:1 mixture of HCl and HNO_3 for approximately 30 to 40 min. Afterwards they were placed into beakers containing ultrapure water and immersed into an ultrasonic cleaner bath for approximately 5 min. The samples were then transferred into quartz tubes containing 6 ml HCl , 1 ml HNO_3 and 6 ml high-purity deionized Milli-Q water ($>18.2 \text{ M}\Omega/\text{cm}$, ELGA water systems). Microwave-assisted acid digestion was performed in a PTFE vessel on a turboWAVE 1500 system (Mikrowellensysteme MWS). The three digestion steps for the microwave are summarized in Table 10.

| Step | t [min] | MW [W] | T [$^{\circ}\text{C}$] | P [bar] |
|------|---------|--------|--------------------------|---------|
| 1 | 5 | 1000 | 180 | 120 |
| 2 | 5 | 1200 | 220 | 150 |
| 3 | 20 | 1200 | 220 | 150 |

Table 10: Microwave oven program for the complete digestion of the sample cuts

4.4.1.3 ICP-MS analysis

A series of test analysis were conducted in order to determine the appropriate dilution factor and operating conditions. The quantitation of major and minor elements requires the application of two separate analyses due to large differences in their concentrations. In contrast to the minor elements, major elements are found in extremely high concentrations. It was determined that the quantitative analysis of the major elements requires dilution with a ratio of 1:250'000, while for the minor elements a dilution ratio of 1:1'000 in 1% HNO_3 was used. This step was performed using a diluter (microLAB 600 series, Hamilton). Internal standards were then added to the samples. In order to eliminate or reduce the effect of interferences, both collision and reaction interface (CRI) gases (He and H_2) were used.

Experimental section

The ICP-MS parameters were based on a method that was initially designed for the analysis of trace metals in drinking water and optimized for our samples¹⁷. Elemental concentrations were then measured with the parameters detailed in Table 11.

| ICP-MS | Aurora M90 (Bruker) | |
|--|--|----------------|
| Spray chamber | Peltier-cooled (3°C), double-pass Scott type | |
| Nebulizer | Quartz MicroMist Low Flow Unifit (0.4 ml·min ⁻¹) | |
| Cones | Nickel | |
| RF Power [kW] | 1.45 | |
| Gas Flow [l·min ⁻¹] | Plasma flow | 18.00 |
| | Auxiliary flow | 1.80 |
| | Nebulizer flow | 1.00 |
| Sample introduction | Sampling depth [mm] | 5 |
| | Pump rate [rpm] | 5 |
| | Stabilization time [s] | 30 |
| Quadrupole scan | Scan mode | Segmented scan |
| | Dwell time [ms] | 10 |
| Attenuation mode | None | |
| Acquisition | Points per peak | 1 |
| | Scans/ Replicate | 30 |
| | Replicates/ Sample | 5 |
| Washing solution | HNO ₃ 1% | |
| Washing time [s] | 10 | |
| Collision reaction interface gas [ml·min ⁻¹] | H ₂ : 90 He: 110 | |

Table 11: Operating conditions for ICP-MS

4.4.2 Method validation

Validation is an essential step in demonstrating that a procedure is suitable for its intended purpose [118] and ensuring that the subsequent analytical results are reliable. In our case, method validation was required to select the variables for the chemical

¹⁷ Method developed by the SCAV at the cantonal laboratory of Vaud

profile. In order to be able to determine a specimen's elemental concentration using ICP-MS and discriminate between specimens based on this information, the measurements must meet certain requirements [119]. First of all, the element's concentration must be above the detection limit of the instrument. Furthermore, they should be homogeneously distributed in the alloy and not affected by spectral interferences.

In order to prevent contamination, the specimens were always handled carefully, all labware was systematically cleaned with diluted nitric acid and only new plasticware was used. Several quality assurance procedures were carried out and during this process several elements, isotopes or CRI gas conditions were removed, to obtain the most reliable data for the construction of the chemical profiles.

The quality procedures for the validation process included:

- Analysis of blank samples (0.1% HNO₃ in UPH₂O + internal standard)
- Analysis of procedure blanks (all the reagents were used in the same concentration as for the preparation of the specimens that underwent the same procedure, including microwave digestion and dilution)
- Repeated analysis of the same specimen (M20) at given intervals during the entire period of ICP-MS analysis in order to assess repeatability
- Analysis of calibration standards to assess the calibration curve for quantification
- Quality control consisting of certified reference materials

Table 12 lists all chemical elements, isotopes, CRI gases and ranges for calibration standards that were measured during the ICP-MS experiments.

Experimental section

| | Element | Isotopes | CRI gas | Calibration standards [ppb] | | | | |
|----------------|-----------------|---------------|-------------------|-----------------------------|--------|--------|--------|--------|
| | | | | Std. 1 | Std. 2 | Std. 3 | Std. 4 | Std. 5 |
| Major elements | Chrome (Cr) | 52 | He/H ₂ | 2 | 10 | 20 | 30 | 40 |
| | Iron (Fe) | 54, 56 | He/H ₂ | 10 | 50 | 100 | 150 | 200 |
| | Nickel (Ni) | 58, 60 | He/H ₂ | 1 | 5 | 10 | 15 | 20 |
| | Aluminum (Al) | 27 | He/H ₂ | 1 | 5 | 10 | 15 | 20 |
| | Silicon (Si) | 28 | H ₂ | 100 | 500 | 1000 | 1500 | 2000 |
| | Phosphorus (P) | 31 | He | 50 | 250 | 500 | 750 | 1000 |
| Minor elements | Titanium (Ti) | 48, 49 | He/H ₂ | 1 | 5 | 10 | 15 | 20 |
| | Vanadium (V) | 51 | He/H ₂ | 20 | 100 | 200 | 300 | 400 |
| | Manganese (Mg) | 55 | He/H ₂ | 400 | 2000 | 4000 | 6000 | 8000 |
| | Cobalt (Co) | 59 | He/H ₂ | 100 | 500 | 1000 | 1500 | 2000 |
| | Copper (Cu) | 63, 65 | He/H ₂ | 20 | 100 | 200 | 300 | 400 |
| | Zinc (Zn) | 66 | He | 0.5 | 2.5 | 5 | 7.5 | 10 |
| | Arsenic (As) | 75 | He/H ₂ | 1 | 5 | 10 | 15 | 20 |
| | Molybdenum (Mo) | 95, 98 | He/H ₂ | 50 | 250 | 500 | 750 | 1000 |
| | Cadmium (Cd) | 112, 114 | He/H ₂ | 0.5 | 2.5 | 5 | 7.5 | 10 |
| | Tin (Sn) | 118 | He/H ₂ | 0.5 | 2.5 | 5 | 7.5 | 10 |
| | Tungsten (W) | 182, 183 | He/H ₂ | 2 | 10 | 20 | 30 | 40 |
| | Lead (Pb) | 206, 207, 208 | He/H ₂ | 0.5 | 2.5 | 5 | 7.5 | 10 |
| | Uranium (U) | 238 | He/H ₂ | 0.5 | 2.5 | 5 | 7.5 | 10 |

Table 12: Overview of elements, isotopes, CRI gas and calibration standards

The major and minor elemental analyses were undertaken in two separate series. Most of the isotopes were analysed using both, He and H₂, to decrease possible interferences (as explained below). Due to analytical difficulties, it was not possible to determine the calibration curves for phosphorus (P) and silicon (S) and these elements were, therefore, no longer considered.

Since the target elements (except for the major elements) are at trace level concentrations, it is crucial to demonstrate that the analytical method can reliably quantify trace element analytes. The typical validation characteristics are: specificity, linearity, accuracy, precision, quantitation and the detection limit [120]. Other characteristics can also be taken into account, depending on the level of validation needed for the purpose of the analytical method.

4.4.2.1 Specificity/ Selectivity

Specificity can be defined as “*the ability to assess unequivocally the analyte in the presence of components which may be expected to be present. Typically these might include impurities, degradants, matrix etc.*” [118]. However, the IUPAC discourages the use of the term ‘specificity’ and recommends the use of the term ‘selectivity’. A definition is for example: “*Selectivity of a method refers to the extent to which it can determine particular analyte(s) in a complex mixture without interference from other components in the mixtures*“ [121].

In ICP-MS, interferences can be classified into three categories: spectral, matrix and physical [122]. There are several potential ways to reduce these interferences. Spectral interferences are generally isobaric (due to isotopes from other elements with the same mass) or polyatomic. Isobaric interferences are compensated for using mathematical correction methods [122], and in this work, equations were applied to minimize isobaric interferences from the matrix. The collision reaction interface (CRI) technology reduces polyatomic interferences by injecting reactive/collision gases into the plasma and therefore preventing interfering ions from entering the mass analyser. In our case, hydrogen (H_2) and helium (He) were used as the CRI gases to reduce polyatomic molecules. Helium was used for collisional interference attenuation. Since polyatomic ions are physically larger than the analyte ions, they collide more frequently with the CRI gas molecules, which causes them to slow down. Consequently, they don’t have enough energy (velocity) left to pass through the ion optics and the mass analyser. This process is called kinetic energy discrimination. Using H_2 , polyatomic molecules are converted into one or more ions with masses that are different from the analyte isotope. Figure 11 shows an example of the molecule $ArCl$, having the same mass as As, and its transformation after reaction with the H_2 CRI gas:

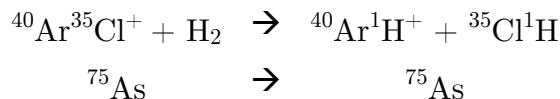


Figure 11: Example of a reactive interference attenuation using H_2

Physical interferences can occur due to viscosity changes in the sample, however, this should not be a problem in our system. All samples were considerably diluted,

decreasing the acidification of the sample and, therefore, matrix effects associated with viscosity changes.

Matrix suppression is caused when the sample affects the ionization and can be reduced using internal standardization. Elements chosen for the internal standard are presumed to not be present in our specimens (nonanalyte isotopes) and cover the mass spectrum. The elements are scandium (Sc), germanium (Ge), yttrium (Y), rhodium (Rh), indium (In), bismuth (Bi) and terbium (Tb). Internal standards are used as a correction factor for matrix effects (loss or gain of the analytical signal), since any variation in the response is corrected for in the analytical results [122]. Monitoring the blank reagent can indicate the presence of contamination from the laboratory environment.

The selectivity can be evaluated by analysing various blank samples, and the acceptance standard is the absence of any interfering signals [123]. Ratios between the mean value of the sample signals for the 3 major and the 14 minor elements and the two blank samples (calibration blank and procedure blank) are shown in Table 13 and Table 14, respectively. In total, six different calibration blanks and two procedure blanks were analysed.

| Element | S mean [cps] | B mean [cps] | PB mean [cps] | Ratio B/S | Ratio PB/S |
|---------|--------------|--------------|---------------|-----------|------------|
| Cr52 | 66656 | 1031 | 75 | 0.0155 | 0.0011 |
| Fe54 | 37627 | 2991 | 2695 | 0.0795 | 0.0716 |
| Fe56 | 610862 | 14737 | 4600 | 0.0241 | 0.0075 |
| Ni58 | 22751 | 989 | 885 | 0.0435 | 0.0389 |
| Ni60 | 16258 | 664 | 579 | 0.0408 | 0.0356 |

Table 13: Mean values in counts per second (cps) for sample signals (S), calibration blanks (B) and procedure blank (PB) and the ratios between sample and blank signals for the three major elements

Experimental section

| Element | S mean [cps] | B mean [cps] | PB mean [cps] | Ratio B/S | Ratio PB/S |
|---------|--------------|--------------|---------------|-----------|------------|
| Al27 | 4361 | 773 | 462 | 0.177 | 0.106 |
| Ti48 | 2193 | -3 | 20 | -0.001 | 0.009 |
| Ti49 | 200 | 3 | 31 | 0.016 | 0.156 |
| V51 | 184013 | 22 | 1848 | 0.000 | 0.010 |
| Mn55 | 138662 | 11 | 6 | 0.000 | 0.000 |
| Co59 | 12092 | 0 | 0 | 0.000 | 0.000 |
| Cu63 | 108443 | 85 | 93 | 0.001 | 0.001 |
| Cu65 | 53751 | 48 | 46 | 0.001 | 0.001 |
| Zn66 | 492 | 148 | 136 | 0.300 | 0.277 |
| As75 | 453 | 40 | 20 | 0.088 | 0.044 |
| Mo95 | 2800 | 1 | 1 | 0.000 | 0.000 |
| Mo98 | 48567 | 7 | 28 | 0.000 | 0.001 |
| Cd112 | -69 | 2 | 2 | -0.033 | -0.032 |
| Cd114 | -82 | 9 | -2 | -0.112 | 0.029 |
| Sn118 | 12054 | 27 | 28 | 0.002 | 0.002 |
| W182 | 41378 | 149 | 217 | 0.004 | 0.005 |
| W183 | 23026 | 83 | 130 | 0.004 | 0.006 |
| Pb206 | 1640 | 175 | 175 | 0.107 | 0.107 |
| Pb207 | 1196 | 159 | 166 | 0.133 | 0.139 |
| Pb208 | 2950 | 377 | 392 | 0.128 | 0.133 |
| U238 | 130 | 6 | 4 | 0.049 | 0.034 |

Table 14: Mean values in counts per second (cps) for sample signals (S), calibration blanks (B) and procedure blank (PB) and the ratios between sample and blank signals for the major elements

In this approach selectivity is demonstrated by the absence of interfering signals. The ratios demonstrate the lack of response in the blank matrix, which shows that external signals do not interfere with the analyte signals. The low ratios between the procedure blanks and the sample signal average also indicates the absence of contamination during specimen preparation and from the laboratory environment.

A few negative values can be seen in Table 14, it is evidently not possible to measure a negative signal for the counts per seconds (cps), and the obtained values are due to the correction equations applied to all elements with potential isobaric interferences. For example, a mass of 48 has two isotopes, Ti (abundance = 73,72%) and Ca

(abundance = 0.187%). In order to measure Ti, a correction equation is applied to subtract the Ca fraction, causing a negative value.

4.4.2.2 Accuracy and Precision

Accuracy (also called bias or trueness) expresses “*the closeness of agreement between the value which is accepted either as a conventional true value or an accepted reference value and the value found*”. It is generally expressed in terms of recovery that should lie within 80% and 120% [120]. In order to assess recovery, experiments using certified reference solutions in the mid-region of the working range were carried out.

Precision is a concept that is usually expressed through repeatability, intermediate repeatability and reproducibility of the measured results [118]. In our case, the precision was estimated by measuring the repeatability of the ICP-MS method based on a certified standard solution and a specimen (M20) that were analysed sequentially. We assessed that the reproducibility of our measurements was not a factor, since the measurements were carried out by the same operator with the same equipment. Nevertheless, the reproducibility is important if a method is intended to be used across different laboratories. Repeatability is defined as the “*closeness of agreement between the test results obtained under the same operating conditions (same laboratory, same equipment, same operator) over a short period of time*” [118]. Usually, repeatability is measured in terms of the relative standard deviation (%RSD), and is therefore not related to reference values. It is generally accepted that the precision at each concentration level should not exceed 15% [124]. In ICP-MS, the standard deviations can be low (with a stable nebulizer gas flow conditions) [119]. This can be seen in Table 15 and Table 16 for the quality control samples.

Experimental section

| Element | Certified conc. [ppb] | Mean measured conc. [ppb] | Recovery [%] | SD [ppb] | %RSD | # Meas. | Isotopes | Selected |
|---------------------|-----------------------|---------------------------|--------------|----------|------|---------|----------|----------|
| Cr(He) | 20 | 20.54 | 102.68 | 0.69 | 3.38 | 28 | 52 | ✓ |
| Cr(H ₂) | 20 | 20.04 | 100.19 | 0.50 | 2.48 | 28 | 52 | ✓ |
| Fe(He) | 100 | 102.72 | 102.72 | 3.17 | 3.09 | 56 | 54, 56 | ✓ |
| Fe(H ₂) | 100 | 101.49 | 101.49 | 2.05 | 2.02 | 56 | 54, 56 | ✓ |
| Ni(He) | 10 | 10.25 | 102.55 | 0.34 | 3.32 | 56 | 58, 60 | ✓ |
| Ni(H ₂) | 10 | 10.20 | 101.96 | 0.24 | 2.34 | 28 | 58, 60 | ✓ |

Table 15: Recovery percentages between the measured (recovered) and certified value and statistical parameters for the major elements. The selection refers to the elements that were retained for the construction of the chemical profile.

For all the major elements and gas conditions, recovery is close to 100% and the %RSD is very low indicating good accuracy and precision (repeatability).

Experimental section

| Element | Certified conc. [ppb] | Mean measured conc. [ppb] | Recovery [%] | SD [ppb] | %RSD | # Meas. | Isotopes | Selected |
|---------------------|-----------------------|---------------------------|--------------|----------|-------|---------|---------------|----------|
| Al(He) | 10 | 7.74 | 77.37 | 1.40 | 13.96 | 24 | 27 | ✗ |
| Al(H ₂) | 10 | 9.00 | 90.00 | 0.97 | 9.67 | 24 | 27 | ✓ |
| Ti(He) | 10 | 10.23 | 102.28 | 0.55 | 5.47 | 48 | 48, 49 | ✓ |
| Ti(H ₂) | 10 | 8.10 | 81.04 | 0.88 | 8.79 | 48 | 48, 49 | ✗ |
| V(He) | 200 | 214.90 | 107.45 | 10.86 | 5.43 | 24 | 51 | ✓ |
| V(H ₂) | 200 | 187.40 | 93.70 | 5.86 | 2.93 | 24 | 51 | ✓ |
| Mn(He) | 4000 | 4225.85 | 105.65 | 182.10 | 4.55 | 24 | 55 | ✓ |
| Mn(H ₂) | 4000 | 4029.53 | 100.74 | 157.21 | 3.93 | 24 | 55 | ✓ |
| Co(He) | 1000 | 1177.13 | 117.71 | 101.67 | 10.17 | 24 | 59 | ✗ |
| Co(H ₂) | 1000 | 984.47 | 98.45 | 27.65 | 2.76 | 24 | 59 | ✓ |
| Cu(He) | 200 | 197.04 | 98.52 | 9.34 | 4.67 | 48 | 63, 65 | ✓ |
| Cu(H ₂) | 200 | 163.27 | 81.63 | 18.62 | 9.31 | 48 | 63, 65 | ✗ |
| Zn(He) | 5 | 5.38 | 107.55 | 0.34 | 6.75 | 24 | 66 | ✓ |
| As(He) | 10 | 10.05 | 100.46 | 0.43 | 4.28 | 24 | 75 | ✓ |
| As(H ₂) | 10 | 10.30 | 103.02 | 0.47 | 4.67 | 24 | 75 | ✓ |
| Mo(He) | 500 | 584.61 | 116.92 | 46.09 | 9.22 | 48 | 95, 98 | ✗ |
| Mo(H ₂) | 500 | 390.98 | 78.20 | 47.07 | 9.41 | 48 | 95, 98 | ✗ |
| Cd(He) | 5 | 5.28 | 105.60 | 0.22 | 4.32 | 48 | 112, 114 | ✓ |
| Cd(H ₂) | 5 | 4.79 | 95.78 | 0.17 | 3.43 | 48 | 112, 114 | ✓ |
| Sn(He) | 5 | 5.07 | 101.45 | 0.14 | 2.78 | 24 | 118 | ✓ |
| Sn(H ₂) | 5 | 4.98 | 99.58 | 0.15 | 2.96 | 24 | 118 | ✓ |
| W(He) | 20 | 19.47 | 97.35 | 0.82 | 4.11 | 48 | 182, 183 | ✓ |
| W(H ₂) | 20 | 18.00 | 90.00 | 0.98 | 4.91 | 48 | 182, 183 | ✓ |
| Pb(He) | 5 | 4.89 | 97.86 | 0.17 | 3.39 | 72 | 206, 207, 208 | ✓ |
| Pb(H ₂) | 5 | 4.91 | 98.26 | 0.13 | 2.57 | 72 | 206, 207, 208 | ✓ |
| U(He) | 5 | 5.10 | 102.05 | 0.13 | 2.61 | 24 | 238 | ✓ |
| U(H ₂) | 5 | 4.99 | 99.74 | 0.15 | 3.02 | 24 | 238 | ✓ |

Table 16: Recovery percentages between the measured (recovered) and certified value and statistical parameters for the minor elements. The selection refers to the elements that were retained for the construction of the chemical profile.

The recovery for Al analysed using He, and Mo using H₂ are below 80% and were therefore eliminated. Some CRI gas conditions were eliminated due to the elevated relative standard deviations (%RSD) compared to the same isotope under condition with the second gas (shown in Table 16). The eliminated conditions and elements were not considered further for the validation procedure or for the chemical profiles. Nevertheless, no choice between isotopes of the same element was made at this point. This decision was taken at a later stage, when the variable selection for construction of the chemical profile of the minor elements was needed.

Since certified calibration solutions were used to study the accuracy of the measurements, some questions will remain unanswered. As we had no access to the solid reference material that went through the entire specimen preparation process, it is not possible to assert whether the dissolution was complete or not. The blank samples were spiked with certified standard solutions, therefore, estimating the matrix effect (changes in the analytical signal) was not possible since real samples had not been spiked. Nonetheless, the internal standardization was used to correct for the matrix effects. In addition, all samples were considerably diluted (1:250'000 for major elements; 1:1'000 for trace elements), decreasing their acidification, and therefore the matrix effects associated with viscosity changes.

Recovery was nearly 100% (between 77% and 117%), indicating minimal losses, good alignment between the standard and the calibration solution and a proper functioning of the analytical system. The %RSDs were low, which is an indication of good precision and that the recovery is stable.

A better estimation of the accuracy and precision could be gained by analysing more than one concentration. Generally, it is recommended that quality control samples include not only the middle portion, but also the lower and upper limit of the calibration range [123]. It would also be possible to establish an accuracy profile; however, it is difficult to find suitable certified reference materials.

Table 17 and Table 18 summarises the values obtained for the repeatability study for specimen M20.

Experimental section

| Element | Mean [g/kg] | SD [g/kg] | %RSD | # Meas. | Isotopes |
|---------|-------------|-----------|------|---------|----------|
| Cr(He) | 177.31 | 3.92 | 2.21 | 9 | 52 |
| Cr(H2) | 171.45 | 3.63 | 2.12 | 9 | 52 |
| Fe(He) | 704.24 | 13.39 | 1.90 | 18 | 54,56 |
| Fe(H2) | 692.01 | 6.09 | 0.88 | 18 | 54,56 |
| Ni(He) | 80.03 | 2.43 | 3.04 | 18 | 58,60 |
| Ni(H2) | 79.29 | 2.10 | 2.64 | 9 | 58,60 |

Table 17: Repeatability values of specimen M20 for the major elements

| Element | Mean [ppb] | SD [ppb] | %RSD | # Meas. | Isotopes |
|---------|------------|----------|--|---------|----------|
| Al(H2) | 6.75 | 1.01 | 14.89 | 4 | 27 |
| Ti(He) | 25.02 | 2.88 | 11.50 | 8 | 48, 49 |
| V(He) | 976.19 | 20.68 | 2.12 | 4 | 51 |
| V(H2) | 834.12 | 17.21 | 2.06 | 4 | 51 |
| Mn(He) | 13698.62 | 275.93 | 2.01 | 4 | 55 |
| Mn(H2) | 13040.36 | 457.22 | 3.51 | 4 | 55 |
| Co(H2) | 1881.02 | 45.70 | 2.43 | 4 | 59 |
| Cu(He) | 222.45 | 8.34 | 3.75 | 8 | 63, 65 |
| Zn(He) | 3.81 | 2.52 | 66.08 X | 4 | 66 |
| As(He) | 14.46 | 1.28 | 8.84 | 4 | 75 |
| As(H2) | 16.17 | 1.52 | 9.42 | 4 | 75 |
| Sn(He) | 5.91 | 0.06 | 1.04 | 4 | 118 |
| Sn(H2) | 5.86 | 0.17 | 2.83 | 4 | 118 |
| W(He) | 26.06 | 1.32 | 5.06 | 8 | 182, 183 |
| W(H2) | 21.74 | 0.50 | 2.28 | 8 | 182, 183 |

Table 18: Repeatability values of specimen M20 for the minor elements

For M20, the repeatability is only shown for elements that are present in the specimen. Zn showed very poor repeatability (66 %RSD), it was therefore excluded from further validation and from the chemical profile of the watchcases.

4.4.2.3 Linearity

Linearity is defined as “the ability (within a given range) to obtain test results which are directly proportional to the concentration (amount) of analyte in the sample” [118].

A linear regression model is usually assumed but has to be verified during the validation step. The linear calibration takes the general form:

$$y = ax + b$$

Usually, linearity is assumed if the coefficient of correlation is $r \geq 0.990$ [125].

Correlation coefficients were calculated using the following equation:

$$r = \frac{\sum(x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum(x_i - \bar{x})^2 \sum(y_i - \bar{y})^2}}$$

Correlation on its own is not a proof of linearity. Other parameters from the regression line, like the slope, y-intercept and the residual sum of the squares should also be reported [118]. The residual sum of the squares is the main estimate of the disparity between the data and the estimation model. The slope of the regression line is correlated with the sensitivity. The stronger the instrument's response on y to a concentration change on x, the steeper the slope and the more sensitive the procedure. The y-intercept provides an estimate of the variability of the method as a signal of the standard blank. It is generally recommended to use at least 5 concentration levels. Moreover, it is suggested that the relative standard deviation of the slope should not exceed 8% for MS-detection based techniques. In addition, several replicates of the calibration curve under repeatable conditions should be taken into account [126].

Five different concentration levels and a blank were used for the calibration curve. The major elements were analysed in 5 series and the minor elements in 4 series. Table 19 summarises the different parameters used to evaluate linearity.

Experimental section

| Major Elements | | | | | | | | | | |
|---|----------|-----------------------|----------|-----------------------|----------|-----------------------|-----------|-----------------------|----------|---------|
| | Cr52(He) | Cr52(H ₂) | Fe54(He) | Fe54(H ₂) | Fe56(He) | Fe56(H ₂) | Ni58(He) | Ni58(H ₂) | Ni60(He) | |
| Slope, <i>a</i> | 1 | 7494.29 | 3428.27 | 259.78 | 1125.33 | 4407.33 | 19073.88 | 6504.53 | 1202.90 | 2750.29 |
| | 2 | 6608.54 | 2970.81 | 227.61 | 1011.36 | 3915.78 | 17141.00 | 5754.23 | 1035.62 | 2458.53 |
| | 3 | 5603.47 | 2485.84 | 190.42 | 835.68 | 3237.82 | 14268.25 | 4827.69 | 831.94 | 2042.83 |
| | 4 | 5235.06 | 2146.26 | 177.71 | 762.08 | 3038.94 | 12930.57 | 4513.67 | 740.69 | 1928.32 |
| | 5 | 5566.52 | 2371.42 | 190.88 | 845.23 | 3297.81 | 14832.45 | 4962.40 | 893.45 | 2116.72 |
| SD of slope, <i>SD_a</i> | 1 | 27.14 | 14.24 | 1.00 | 2.05 | 17.38 | 64.68 | 13.30 | 5.74 | 14.65 |
| | 2 | 15.76 | 20.31 | 0.75 | 5.59 | 11.04 | 45.96 | 18.14 | 7.20 | 9.60 |
| | 3 | 35.09 | 5.69 | 1.34 | 1.65 | 15.11 | 26.28 | 25.49 | 10.78 | 13.70 |
| | 4 | 13.22 | 9.80 | 1.10 | 0.96 | 7.47 | 62.64 | 8.14 | 4.09 | 2.45 |
| | 5 | 279.73 | 185.99 | 10.03 | 67.15 | 203.98 | 1469.49 | 326.69 | 108.42 | 136.98 |
| RSD slope, <i>RSD_a</i> | 1 | 0.36 | 0.42 | 0.38 | 0.18 | 0.39 | 0.34 | 0.20 | 0.48 | 0.53 |
| | 2 | 0.24 | 0.68 | 0.33 | 0.55 | 0.28 | 0.27 | 0.32 | 0.69 | 0.39 |
| | 3 | 0.63 | 0.23 | 0.71 | 0.20 | 0.47 | 0.18 | 0.53 | 1.30 | 0.67 |
| | 4 | 0.25 | 0.46 | 0.62 | 0.13 | 0.25 | 0.48 | 0.18 | 0.55 | 0.13 |
| | 5 | 5.03 | 7.84 | 5.26 | 7.94 | 6.19 | 9.91 | 6.58 | 12.13 | 6.47 |
| y-intercept, <i>b</i> | 1 | 525.53 | 18.59 | 1090.94 | 5570.72 | 11160.13 | 14412.47 | 1554.81 | 328.91 | 659.35 |
| | 2 | 357.76 | -326.90 | 1016.63 | 4267.04 | 8442.24 | 3879.82 | 1194.46 | 324.35 | 483.32 |
| | 3 | -271.99 | 77.43 | 744.17 | 3596.48 | 8196.42 | 5469.02 | 982.94 | 257.49 | 369.57 |
| | 4 | 372.29 | -64.26 | 707.98 | 3802.00 | 7535.82 | 12187.94 | 1281.02 | 350.55 | 527.52 |
| | 5 | 1162.49 | 371.82 | 670.48 | 4367.33 | 8185.64 | 12885.08 | 879.93 | 164.54 | 344.50 |
| <i>r</i> | 1 | 0.99997 | 0.99997 | 0.99997 | 0.99999 | 0.99997 | 0.99998 | 0.99999 | 0.99995 | 0.99994 |
| | 2 | 0.99999 | 0.99991 | 0.99998 | 0.99994 | 0.99998 | 0.99999 | 0.99998 | 0.99990 | 0.99997 |
| | 3 | 0.99992 | 0.99999 | 0.99990 | 0.99999 | 0.99996 | 0.99999 | 0.99994 | 0.99966 | 0.99991 |
| | 4 | 0.99999 | 0.99996 | 0.99992 | 1.00000 | 0.99999 | 0.99995 | 0.99999 | 0.99994 | 1.00000 |
| | 5 | 0.99674 | 0.99230 | 0.99687 | 0.99230 | 0.99546 | 0.98878 | 0.99545 | 0.98549 | 0.99561 |
| <i>r</i> ² | 1 | 0.99995 | 0.99993 | 0.99994 | 0.99999 | 0.99994 | 0.99995 | 0.99998 | 0.99991 | 0.99989 |
| | 2 | 0.99998 | 0.99981 | 0.99996 | 0.99988 | 0.99997 | 0.99997 | 0.99996 | 0.99981 | 0.99994 |
| | 3 | 0.99984 | 0.99998 | 0.99980 | 0.99998 | 0.99991 | 0.99999 | 0.99989 | 0.99933 | 0.99982 |
| | 4 | 0.99997 | 0.99992 | 0.99985 | 0.99999 | 0.99998 | 0.99991 | 0.99999 | 0.99988 | 0.99999 |
| | 5 | 0.99349 | 0.98465 | 0.99374 | 0.98467 | 0.99094 | 0.97769 | 0.99092 | 0.97119 | 0.99125 |
| residual SD curve, <i>SD_{res}</i> | 1 | 967.24 | 507.39 | 177.97 | 364.44 | 3097.11 | 11524.76 | 236.92 | 102.25 | 261.02 |
| | 2 | 561.77 | 723.87 | 133.80 | 996.35 | 1967.40 | 8189.27 | 323.22 | 128.23 | 171.05 |
| | 3 | 1250.44 | 202.65 | 239.63 | 293.23 | 2692.92 | 4683.04 | 454.20 | 192.06 | 244.10 |
| | 4 | 471.17 | 349.21 | 195.99 | 171.91 | 1331.81 | 11162.40 | 144.97 | 72.88 | 43.58 |
| | 5 | 9968.91 | 6628.22 | 1787.80 | 11965.17 | 36345.60 | 261841.80 | 5821.07 | 1931.82 | 2440.71 |

Table 19: Linear regression parameters for the analysed isotopes of the major elements during 5 series of analysis, where «r» refers to the correlation coefficient and «r²» to the determination coefficient

From Table 19 it can be seen, that the determination coefficients were high (the lowest value being 0.971). The values for %RSD_a were very low and exceeded the critical 8% only for Fe56 (H₂) and for Ni58 (H₂) in one series. Therefore, the linearity for the major elements was considered satisfactory. For linear relationships, the slope is correlated with the sensitivity (the greater the change of signal from one concentration to the next one, the higher the sensitivity). Hence, the measurements were the most sensitive for ⁵²Cr using He, ⁵⁶Fe using He and ⁵⁸Ni using H₂. This is not surprising since they are the primary isotopes for each element but still useful for choosing the CRI gas conditions.

Experimental section

| | | Minor Elements | | | | | | | | |
|----------------------------------|---|----------------|----------|----------|----------|----------|----------|----------|----------|----------|
| | | Al27(He) | Ti48(He) | Ti49(He) | V51(He) | V51(H2) | Mn55(He) | Mn55(H2) | Co59(H2) | Cu63(He) |
| Slope, a | 1 | 4324.15 | 933.01 | 74.10 | 2296.72 | 2159.30 | 22.02 | 189.87 | 45.62 | 3987.94 |
| | 2 | 4269.40 | 1038.50 | 80.91 | 2535.41 | 2288.07 | 24.87 | 193.70 | 48.72 | 4411.27 |
| | 3 | 3125.90 | 654.44 | 102.43 | 1686.12 | 1666.97 | 16.13 | 148.82 | 36.22 | 3132.52 |
| | 4 | 12596.83 | 2309.67 | 184.28 | 5372.65 | 5360.05 | 53.57 | 456.22 | 112.10 | 8508.77 |
| SD of slope, SD_a | 1 | 248.82 | 57.57 | 3.78 | 136.97 | 108.52 | 1.38 | 9.06 | 2.20 | 226.98 |
| | 2 | 302.77 | 49.47 | 5.11 | 109.27 | 168.47 | 1.07 | 13.84 | 3.17 | 171.13 |
| | 3 | 72.34 | 23.96 | 3.86 | 62.04 | 41.44 | 0.58 | 3.63 | 0.84 | 115.30 |
| | 4 | 201.75 | 36.74 | 2.66 | 85.37 | 81.24 | 0.79 | 7.96 | 1.45 | 141.65 |
| RSD slope, RSD_a | 1 | 5.75 | 6.17 | 5.10 | 5.96 | 5.03 | 6.27 | 4.77 | 4.83 | 5.69 |
| | 2 | 7.09 | 4.76 | 6.32 | 4.31 | 7.36 | 4.28 | 7.14 | 6.51 | 3.88 |
| | 3 | 2.31 | 3.66 | 3.77 | 3.68 | 2.49 | 3.63 | 2.44 | 2.33 | 3.68 |
| | 4 | 1.60 | 1.59 | 1.44 | 1.59 | 1.52 | 1.47 | 1.75 | 1.30 | 1.66 |
| y-intercept, b | 1 | 5680.02 | 510.63 | 57.93 | 33065.75 | 29489.45 | 6468.71 | 49178.28 | 2867.33 | 55934.62 |
| | 2 | 5911.42 | 357.88 | 44.96 | 23208.48 | 36755.75 | 4505.57 | 60676.90 | 3474.44 | 39296.07 |
| | 3 | 1893.05 | 231.35 | 37.39 | 12310.05 | 10103.90 | 2339.41 | 18113.98 | 1033.48 | 24912.56 |
| | 4 | 9480.72 | 88.04 | 47.26 | 23626.12 | 24324.33 | 4689.55 | 49031.20 | 2368.09 | 45214.92 |
| r | 1 | 0.99344 | 0.99247 | 0.99485 | 0.99296 | 0.99499 | 0.99223 | 0.99547 | 0.99537 | 0.99358 |
| | 2 | 0.99009 | 0.99549 | 0.99211 | 0.99631 | 0.98933 | 0.99635 | 0.98994 | 0.99163 | 0.99700 |
| | 3 | 0.99893 | 0.99733 | 0.99717 | 0.99730 | 0.99877 | 0.99738 | 0.99881 | 0.99892 | 0.99730 |
| | 4 | 0.99949 | 0.99949 | 0.99958 | 0.99950 | 0.99954 | 0.99957 | 0.99939 | 0.99966 | 0.99945 |
| r^2 | 1 | 0.98693 | 0.98500 | 0.98972 | 0.98597 | 0.99000 | 0.98451 | 0.99097 | 0.99076 | 0.98721 |
| | 2 | 0.98028 | 0.99100 | 0.98428 | 0.99263 | 0.97877 | 0.99272 | 0.97999 | 0.98334 | 0.99402 |
| | 3 | 0.99786 | 0.99467 | 0.99435 | 0.99461 | 0.99753 | 0.99477 | 0.99763 | 0.99784 | 0.99461 |
| | 4 | 0.99898 | 0.99899 | 0.99917 | 0.99899 | 0.99908 | 0.99913 | 0.99878 | 0.99933 | 0.99889 |
| residual SD curve, SD_{res} | 1 | 4433.61 | 1025.83 | 67.29 | 48812.28 | 38672.05 | 9841.54 | 64601.65 | 3925.51 | 80890.26 |
| | 2 | 5394.84 | 881.56 | 91.09 | 38938.93 | 60039.42 | 7590.81 | 98637.82 | 5650.77 | 60985.48 |
| | 3 | 1288.91 | 426.91 | 68.79 | 22107.65 | 14769.08 | 4168.73 | 25862.32 | 1501.87 | 41089.10 |
| | 4 | 3594.81 | 654.59 | 47.43 | 30421.79 | 28953.20 | 5628.11 | 56758.07 | 2590.69 | 50479.80 |

Table 20: Linear regression parameters for the analyzed isotopes of some of the minor elements (Al, Ti, V, Mn, Co and Cu) during 4 series of analysis

Experimental section

| | | Minor Elements | | | | | | | | |
|----------------------------------|---|----------------|----------|----------|----------|----------|----------|-----------|-----------|-----------|
| | | Cu65(He) | Zn66(He) | As75(He) | As75(H2) | Mo95(He) | Mo98(He) | Cd112(He) | Cd114(He) | Cd112(H2) |
| Slope, a | 1 | 1965.26 | 421.84 | 191.40 | 163.52 | 18.69 | 32.75 | 1123.72 | 1389.94 | 7669.16 |
| | 2 | 2164.38 | 491.29 | 217.61 | 176.49 | 20.36 | 35.25 | 1306.20 | 1565.48 | 7773.81 |
| | 3 | 1549.47 | 320.51 | 146.89 | 126.45 | 15.92 | 27.65 | 1003.14 | 1214.71 | 6233.78 |
| | 4 | 4159.34 | 835.48 | 381.79 | 343.58 | 34.94 | 59.79 | 1999.33 | 2410.65 | 12652.42 |
| SD of slope, SD_a | 1 | 108.37 | 23.50 | 9.65 | 7.75 | 0.84 | 1.28 | 49.09 | 58.25 | 233.50 |
| | 2 | 82.63 | 16.85 | 7.35 | 15.24 | 0.55 | 1.04 | 29.67 | 39.70 | 313.77 |
| | 3 | 55.36 | 10.44 | 4.68 | 3.74 | 0.48 | 0.67 | 24.15 | 33.67 | 104.19 |
| | 4 | 66.39 | 24.48 | 4.57 | 8.61 | 0.44 | 0.81 | 32.69 | 35.26 | 184.15 |
| RSD slope, RSD_a | 1 | 5.51 | 5.57 | 5.04 | 4.74 | 4.49 | 3.90 | 4.37 | 4.19 | 3.04 |
| | 2 | 3.82 | 3.43 | 3.38 | 8.64 | 2.70 | 2.96 | 2.27 | 2.54 | 4.04 |
| | 3 | 3.57 | 3.26 | 3.19 | 2.96 | 3.04 | 2.42 | 2.41 | 2.77 | 1.67 |
| | 4 | 1.60 | 2.93 | 1.20 | 2.51 | 1.25 | 1.35 | 1.64 | 1.46 | 1.46 |
| y-intercept, b | 1 | 26347.21 | 418.34 | 202.32 | 124.28 | 440 | 746.13 | 318.02 | 353.12 | 1353.00 |
| | 2 | 18605.42 | 292.10 | 165.87 | 192.62 | 348 | 630.44 | 159.20 | 265.73 | 1682.54 |
| | 3 | 11956.30 | 144.38 | 110.04 | 58.17 | 243 | 391.30 | 143.80 | 189.93 | 652.97 |
| | 4 | 20700.83 | 526.10 | 216.64 | 102.44 | 334 | 647.37 | 294.08 | 294.46 | 1465.05 |
| r | 1 | 0.99397 | 0.99385 | 0.99496 | 0.99554 | 0.99599 | 0.99697 | 0.99621 | 0.99651 | 0.99815 |
| | 2 | 0.99710 | 0.99766 | 0.99773 | 0.98541 | 0.99855 | 0.99825 | 0.99897 | 0.99872 | 0.99676 |
| | 3 | 0.99746 | 0.99788 | 0.99797 | 0.99826 | 0.99815 | 0.99883 | 0.99884 | 0.99847 | 0.99944 |
| | 4 | 0.99949 | 0.99829 | 0.99971 | 0.99875 | 0.99969 | 0.99963 | 0.99947 | 0.99957 | 0.99958 |
| r^2 | 1 | 0.98798 | 0.98774 | 0.98994 | 0.99110 | 0.99200 | 0.99394 | 0.99242 | 0.99302 | 0.99631 |
| | 2 | 0.99420 | 0.99532 | 0.99546 | 0.97102 | 0.99710 | 0.99651 | 0.99794 | 0.99743 | 0.99353 |
| | 3 | 0.99492 | 0.99577 | 0.99595 | 0.99652 | 0.99631 | 0.99767 | 0.99769 | 0.99694 | 0.99888 |
| | 4 | 0.99898 | 0.99658 | 0.99943 | 0.99749 | 0.99938 | 0.99927 | 0.99893 | 0.99914 | 0.99915 |
| residual SD curve, SD_{res} | 1 | 38618.89 | 209.38 | 171.93 | 138.03 | 747.50 | 1139.20 | 437.33 | 519.00 | 2080.30 |
| | 2 | 29446.85 | 150.10 | 130.89 | 271.63 | 489.28 | 929.50 | 264.34 | 353.70 | 2795.44 |
| | 3 | 19728.11 | 93.03 | 83.46 | 66.58 | 431.75 | 595.67 | 215.15 | 299.93 | 928.21 |
| | 4 | 23659.92 | 218.10 | 81.48 | 153.48 | 387.81 | 720.11 | 291.29 | 314.15 | 1640.60 |

Table 21: Linear regression parameters for the analyzed isotopes of some of the minor elements (Cu, Zn, As, Mo and Cd) during 4 series of analysis

Experimental section

| | | Minor Elements | | | | | | | | |
|----------------------------------|---|----------------|-----------|-----------|----------|----------|----------|----------|-----------|-----------|
| | | Cd114(H2) | Sn118(He) | Sn118(H2) | W182(He) | W183(He) | W182(H2) | W183(H2) | Pb206(He) | Pb207(He) |
| Slope, a | 1 | 9104.89 | 1000.41 | 14545.56 | 9089.75 | 5015.93 | 14349.64 | 9090.00 | 3904.34 | 758.67 |
| | 2 | 9268.59 | 14460.48 | 1158.83 | 9990.89 | 5530.06 | 15096.69 | 9991.00 | 4548.26 | 665.59 |
| | 3 | 7448.41 | 876.02 | 12061.43 | 9107.18 | 5034.47 | 12053.26 | 9107.00 | 4131.60 | 988.64 |
| | 4 | 15003.06 | 1837.91 | 24145.74 | 12215.24 | 6729.89 | 17340.28 | 12215.00 | 5279.36 | 950.53 |
| SD of slope, SD_a | 1 | 312.14 | 43.60 | 459.27 | 170.17 | 88.62 | 251.63 | 170.17 | 85.16 | 74.98 |
| | 2 | 415.39 | 613.68 | 29.88 | 139.53 | 73.92 | 334.30 | 139.53 | 74.71 | 74.40 |
| | 3 | 151.66 | 23.15 | 224.70 | 211.44 | 126.03 | 237.19 | 211.44 | 110.97 | 109.43 |
| | 4 | 262.81 | 38.60 | 369.07 | 266.76 | 144.61 | 277.02 | 266.76 | 106.69 | 100.43 |
| RSD slope, RSD_a | 1 | 3.43 | 4.36 | 3.16 | 1.87 | 1.77 | 1.75 | 1.87 | 2.18 | 9.88 X |
| | 2 | 4.48 | 4.24 | 2.58 | 1.40 | 1.34 | 2.21 | 1.40 | 1.64 | 11.18 X |
| | 3 | 2.04 | 2.64 | 1.86 | 2.32 | 2.50 | 1.97 | 2.32 | 2.69 | 11.07 X |
| | 4 | 1.75 | 2.10 | 1.53 | 2.18 | 2.15 | 1.60 | 2.18 | 2.02 | 10.57 X |
| y-intercept, b | 1 | 1816.53 | 229.43 | 2636.56 | 5022.38 | 2780.69 | 6143.82 | 5022.00 | 641.71 | 523.05 |
| | 2 | 2195.04 | 3475.63 | 180.22 | 4813.82 | 2604.56 | 8216.03 | 4814.00 | 631.21 | 651.84 |
| | 3 | 985.40 | 136.50 | 1387.67 | 5969.76 | 3525.28 | 6139.38 | 5970.00 | 857.72 | 765.36 |
| | 4 | 1864.50 | 293.59 | 3225.16 | 9088.37 | 4864.91 | 9177.44 | 9088.00 | 1215.28 | 1059.87 |
| r | 1 | 0.99766 | 0.99622 | 0.99801 | 0.99930 | 0.99938 | 0.99939 | 0.99930 | 0.99905 | 0.99925 |
| | 2 | 0.99601 | 0.99642 | 0.99867 | 0.99961 | 0.99964 | 0.99902 | 0.99961 | 0.99946 | 0.99945 |
| | 3 | 0.99917 | 0.99861 | 0.99931 | 0.99892 | 0.99875 | 0.99923 | 0.99892 | 0.99856 | 0.99857 |
| | 4 | 0.99939 | 0.99912 | 0.99953 | 0.99905 | 0.99908 | 0.99949 | 0.99905 | 0.99918 | 0.99927 |
| r^2 | 1 | 0.99532 | 0.99246 | 0.99603 | 0.99860 | 0.99875 | 0.99877 | 0.99860 | 0.99810 | 0.99850 |
| | 2 | 0.99203 | 0.99285 | 0.99735 | 0.99922 | 0.99929 | 0.99804 | 0.99922 | 0.99892 | 0.99889 |
| | 3 | 0.99834 | 0.99721 | 0.99861 | 0.99785 | 0.99750 | 0.99845 | 0.99785 | 0.99712 | 0.99713 |
| | 4 | 0.99877 | 0.99824 | 0.99907 | 0.99810 | 0.99816 | 0.99898 | 0.99810 | 0.99837 | 0.99853 |
| residual SD curve, SD_{res} | 1 | 2780.90 | 388.41 | 4091.74 | 6064.34 | 3158.24 | 8967.49 | 6064.34 | 758.67 | 668.03 |
| | 2 | 3700.78 | 5467.46 | 266.17 | 4972.26 | 2634.37 | 11913.57 | 4972.26 | 665.59 | 662.87 |
| | 3 | 1351.22 | 206.23 | 2001.90 | 7535.02 | 4491.23 | 8452.90 | 7535.02 | 988.64 | 974.91 |
| | 4 | 2341.49 | 343.85 | 3288.11 | 9506.68 | 5153.44 | 9872.27 | 9506.68 | 950.53 | 894.74 |

Table 22: Linear regression parameters for the analyzed isotopes of some of the minor elements (Cd, Sn, W and Pb) during 4 series of analysis

Experimental section

| | | Minor Elements | | | | | |
|---|---|----------------|------------------------|------------------------|------------------------|----------|-----------------------|
| | | Pb208(He) | Pb206(H ₂) | Pb207(H ₂) | Pb208(H ₂) | U238(He) | U238(H ₂) |
| Slope, <i>a</i> | 1 | 9164.81 | 29892.67 | 29177.47 | 68910.67 | 20032.48 | 186848.24 |
| | 2 | 10532.02 | 30366.94 | 29569.88 | 69996.29 | 24123.47 | 187942.74 |
| | 3 | 9712.82 | 26916.97 | 26247.37 | 62016.12 | 23027.12 | 175435.05 |
| | 4 | 12305.06 | 36290.58 | 35458.59 | 83373.92 | 27819.17 | 223523.09 |
| SD of slope, <i>SD_a</i> | 1 | 198.55 | 601.80 | 535.43 | 1307.41 | 431.09 | 4068.43 |
| | 2 | 169.53 | 727.44 | 874.68 | 1756.93 | 334.52 | 5509.40 |
| | 3 | 231.69 | 641.56 | 637.30 | 1467.54 | 672.87 | 4366.28 |
| | 4 | 264.75 | 405.00 | 428.19 | 1004.05 | 266.72 | 1921.14 |
| RSD slope, <i>RSD_a</i> | 1 | 2.17 | 2.01 | 1.84 | 1.90 | 2.15 | 2.18 |
| | 2 | 1.61 | 2.40 | 2.96 | 2.51 | 1.39 | 2.93 |
| | 3 | 2.39 | 2.38 | 2.43 | 2.37 | 2.92 | 2.49 |
| | 4 | 2.15 | 1.12 | 1.21 | 1.20 | 0.96 | 0.86 |
| y-intercept, <i>b</i> | 1 | 1383.31 | 3568.67 | 3638.64 | 8084.84 | 2886.24 | 21104.19 |
| | 2 | 1547.86 | 4949.71 | 5275.23 | 10974.41 | 2959.00 | 31011.90 |
| | 3 | 1744.59 | 4121.86 | 4464.86 | 9971.38 | 4618.91 | 27778.79 |
| | 4 | 2784.54 | 4220.32 | 4243.47 | 9659.05 | 2980.98 | 17432.26 |
| <i>r</i> | 1 | 0.99906 | 0.99919 | 0.99933 | 0.99928 | 0.99908 | 0.99905 |
| | 2 | 0.99948 | 0.99885 | 0.99825 | 0.99874 | 0.99962 | 0.99829 |
| | 3 | 0.99886 | 0.99887 | 0.99882 | 0.99888 | 0.99830 | 0.99876 |
| | 4 | 0.99908 | 0.99975 | 0.99971 | 0.99971 | 0.99982 | 0.99985 |
| <i>r</i> ² | 1 | 0.99813 | 0.99838 | 0.99865 | 0.99856 | 0.99815 | 0.99811 |
| | 2 | 0.99896 | 0.99771 | 0.99651 | 0.99749 | 0.99923 | 0.99657 |
| | 3 | 0.99773 | 0.99773 | 0.99765 | 0.99777 | 0.99660 | 0.99753 |
| | 4 | 0.99815 | 0.99950 | 0.99942 | 0.99942 | 0.99963 | 0.99970 |
| residual SD curve, <i>SD_{res}</i> | 1 | 1768.93 | 5361.61 | 4770.31 | 11648.07 | 3840.68 | 36246.70 |
| | 2 | 1510.38 | 6480.96 | 7792.75 | 15652.98 | 2980.36 | 49084.73 |
| | 3 | 2064.16 | 5715.80 | 5677.90 | 13074.69 | 5994.75 | 38900.36 |
| | 4 | 2358.76 | 3608.24 | 3814.83 | 8945.37 | 2376.31 | 17115.93 |

Table 23: Linear regression parameters for the analyzed isotopes of some of the minor elements (Pb and U) during 4 series of analysis

From Table 20, Table 21, Table 22 and Table 23 it can be seen that the determination coefficients were always very high. Almost all values were above 0.99 (lowest value being 0.971) and therefore satisfactory. The values of %RSD_a were low except for Pb207 using He. Regarding sensitivity, Al27 and Co59 are monoisotopic and the most advantageous CRI gas condition was found during the accuracy study (He for Al27, H₂

for Co59). V51 and As75 are equal in terms of sensitivity under both gas conditions. For the remaining elements, as can be seen from the slopes of the calibration curve, the sensitivity was better for Ti48 using He, Mn55 using H₂, Cu63 using He, Cd114 using H₂, Sn118 using H₂, W182 using H₂, P208 using H₂ and U238 using H₂. Some of the elements were not found in any of the specimens or were below the limit of detection and therefore not used for the construction of the chemical profile.

4.4.2.4 Detection and quantification limits

The detection limit of an analytical procedure is defined as “*the lowest amount of analyte in a sample which can be detected but not necessarily quantitated as an exact value*” and the quantification limit “*the lowest amount of analyte in a sample which can be quantitatively determined with suitable precision and accuracy*” [118].

There are several ways to calculate the detection and quantification limits. In our case, the detection and quantification limits were based on the standard deviation of the response and the slope, as proposed by the International Conference of Harmonization [118].

The limit of detection was calculated as follows:

$$LOD = \frac{3.3 s_{res}}{a}$$

and the limit of quantification,

$$LOQ = \frac{10 s_{res}}{a}$$

where s_{res} is the residual standard deviation of the regression line and a the slope of the calibration curve.

Experimental section

| Element | LOD [g/kg] | LOQ [g/kg] | Element | LOD [g/kg] | LOQ [g/kg] |
|----------|------------|------------|----------|------------|------------|
| Cr52(He) | 1 0.11 | 0.32 | Fe56(H2) | 1 0.50 | 1.51 |
| | 2 0.07 | 0.21 | | 2 0.39 | 1.19 |
| | 3 0.18 | 0.56 | | 3 0.27 | 0.82 |
| | 4 0.07 | 0.23 | | 4 0.71 | 2.16 |
| | 5 1.48 | 4.48 | | 5 14.56 | 44.13 |
| Cr52(H2) | 1 0.12 | 0.37 | Ni58(He) | 1 0.03 | 0.09 |
| | 2 0.20 | 0.61 | | 2 0.05 | 0.14 |
| | 3 0.07 | 0.20 | | 3 0.08 | 0.24 |
| | 4 0.13 | 0.41 | | 4 0.03 | 0.08 |
| | 5 2.31 | 6.99 | | 5 0.97 | 2.93 |
| Fe54(He) | 1 0.57 | 1.71 | Ni58(H2) | 1 0.07 | 0.21 |
| | 2 0.48 | 1.47 | | 2 0.10 | 0.31 |
| | 3 1.04 | 3.15 | | 3 0.19 | 0.58 |
| | 4 0.91 | 2.76 | | 4 0.08 | 0.25 |
| | 5 7.73 | 23.42 | | 5 1.78 | 5.41 |
| Fe54(H2) | 1 0.27 | 0.81 | Ni60(He) | 1 0.08 | 0.24 |
| | 2 0.81 | 2.46 | | 2 0.06 | 0.17 |
| | 3 0.29 | 0.88 | | 3 0.10 | 0.30 |
| | 4 0.19 | 0.56 | | 4 0.02 | 0.06 |
| | 5 11.68 | 35.39 | | 5 0.95 | 2.88 |
| Fe56(He) | 1 0.58 | 1.76 | | | |
| | 2 0.41 | 1.26 | | | |
| | 3 0.69 | 2.08 | | | |
| | 4 0.36 | 1.10 | | | |
| | 5 9.09 | 27.55 | | | |

Table 24: Limit of detection (LOD) and limit of quantification (LOQ) for all major elements under different CRI gas conditions during 5 series of analysis

Experimental section

| Element | LOD [mg/kg] | LOQ [mg/kg] | Element | LOD [mg/kg] | LOQ [mg/kg] |
|----------|-------------|-------------|----------|-------------|-------------|
| Al27(H2) | 1 0.34 | 1.03 | Co59(H2) | 1 28.40 | 86.05 |
| | 2 0.42 | 1.26 | | 2 38.27 | 115.98 |
| | 3 0.14 | 0.41 | | 3 13.68 | 41.47 |
| | 4 0.09 | 0.29 | | 4 7.63 | 23.11 |
| Ti48(He) | 1 0.36 | 1.10 | Cu63(He) | 1 6.69 | 20.28 |
| | 2 0.28 | 0.85 | | 2 4.56 | 13.82 |
| | 3 0.22 | 0.65 | | 3 4.33 | 13.12 |
| | 4 0.09 | 0.28 | | 4 1.96 | 5.93 |
| Ti49(He) | 1 0.30 | 0.91 | Cu65(He) | 1 6.48 | 19.65 |
| | 2 0.37 | 1.13 | | 2 4.49 | 13.61 |
| | 3 0.22 | 0.67 | | 3 4.20 | 12.73 |
| | 4 0.08 | 0.26 | | 4 1.88 | 5.69 |
| V51(He) | 1 7.01 | 21.25 | As75(He) | 1 0.30 | 0.90 |
| | 2 5.07 | 15.36 | | 2 0.20 | 0.60 |
| | 3 4.33 | 13.11 | | 3 0.19 | 0.57 |
| | 4 1.87 | 5.66 | | 4 0.07 | 0.21 |
| V51(H2) | 1 5.91 | 17.91 | As75(H2) | 1 0.28 | 0.84 |
| | 2 8.66 | 26.24 | | 2 0.51 | 1.54 |
| | 3 2.92 | 8.86 | | 3 0.17 | 0.53 |
| | 4 1.78 | 5.40 | | 4 0.15 | 0.45 |
| Mn55(He) | 1 147.50 | 446.97 | Mo95(He) | 1 13.20 | 40.00 |
| | 2 100.71 | 305.17 | | 2 7.93 | 24.03 |
| | 3 85.29 | 258.46 | | 3 8.95 | 27.12 |
| | 4 34.67 | 105.06 | | 4 3.66 | 11.10 |
| Mn55(H2) | 1 112.28 | 340.25 | Mo98(He) | 1 11.48 | 34.78 |
| | 2 168.04 | 509.22 | | 2 8.70 | 26.37 |
| | 3 57.35 | 173.78 | | 3 7.11 | 21.54 |
| | 4 41.06 | 124.41 | | 4 3.97 | 12.04 |

Table 25: Limit of detection (LOD) and limit of quantification (LOQ) for some minor elements (Al, Ti, V, Mn, Co, Cu, As and Mo) under different CRI gas conditions during 4 series of analysis

Experimental section

| Element | LOD [mg/kg] | LOQ [mg/kg] | Element | LOD [mg/kg] | LOQ [mg/kg] |
|-----------|-------------|-------------|-----------|-------------|-------------|
| Cd112(He) | 1 0.13 | 0.39 | W183(He) | 1 0.21 | 0.63 |
| | 2 0.07 | 0.20 | | 2 0.16 | 0.48 |
| | 3 0.07 | 0.21 | | 3 0.29 | 0.89 |
| | 4 0.05 | 0.15 | | 4 0.25 | 0.77 |
| Cd114(He) | 1 0.12 | 0.37 | W182(H2) | 1 0.21 | 0.62 |
| | 2 0.07 | 0.23 | | 2 0.26 | 0.79 |
| | 3 0.08 | 0.25 | | 3 0.23 | 0.70 |
| | 4 0.04 | 0.13 | | 4 0.19 | 0.57 |
| Cd112(H2) | 1 0.09 | 0.27 | W183(H2) | 1 0.22 | 0.67 |
| | 2 0.12 | 0.36 | | 2 0.16 | 0.50 |
| | 3 0.05 | 0.15 | | 3 0.27 | 0.83 |
| | 4 0.04 | 0.13 | | 4 0.26 | 0.78 |
| Cd114(H2) | 1 0.10 | 0.31 | Pb206(He) | 1 0.06 | 0.19 |
| | 2 0.13 | 0.40 | | 2 0.05 | 0.15 |
| | 3 0.06 | 0.18 | | 3 0.08 | 0.24 |
| | 4 0.05 | 0.16 | | 4 0.06 | 0.18 |
| Sn118(He) | 1 0.13 | 0.39 | Pb208(He) | 1 0.06 | 0.19 |
| | 2 0.12 | 0.38 | | 2 0.05 | 0.14 |
| | 3 0.08 | 0.24 | | 3 0.07 | 0.21 |
| | 4 0.06 | 0.19 | | 4 0.06 | 0.19 |
| Sn118(H2) | 1 0.09 | 0.28 | Pb206(H2) | 1 0.06 | 0.18 |
| | 2 0.08 | 0.23 | | 2 0.07 | 0.21 |
| | 3 0.05 | 0.17 | | 3 0.07 | 0.21 |
| | 4 0.04 | 0.14 | | 4 0.03 | 0.10 |
| W182(He) | 1 0.22 | 0.67 | Pb207(H2) | 1 0.05 | 0.16 |
| | 2 0.16 | 0.50 | | 2 0.09 | 0.26 |
| | 3 0.27 | 0.83 | | 3 0.07 | 0.22 |
| | 4 0.26 | 0.78 | | 4 0.04 | 0.11 |

Table 26: Limit of detection (LOD) and limit of quantification (LOQ) for some minor elements (Cd, Sn, W and Pb) under different CRI gas conditions during 4 series of analysis

Experimental section

| Element | LOD [mg/kg] | LOQ [mg/kg] |
|-----------|-------------|-------------|
| Pb208(H2) | 1 0.06 | 0.17 |
| | 2 0.07 | 0.22 |
| | 3 0.07 | 0.21 |
| | 4 0.04 | 0.11 |
| U238(He) | 1 0.06 | 0.19 |
| | 2 0.04 | 0.12 |
| | 3 0.09 | 0.26 |
| | 4 0.03 | 0.09 |
| U238(H2) | 1 0.06 | 0.19 |
| | 2 0.09 | 0.26 |
| | 3 0.07 | 0.22 |
| | 4 0.03 | 0.08 |

Table 27: Limit of detection (LOD) and limit of quantification (LOQ) for some minor elements (Pb and U) under different CRI gas conditions during 4 series of analysis

4.4.2.5 Summary of the method validation

During the validation process, some elements and isotopes, as well as certain CRI gas conditions, were eliminated due to non-fulfilment of the acceptance criteria. The validated elements and CRI gas conditions are shown in Table 28 and Table 29:

| Validated major elements | |
|--------------------------|----------|
| Cr52(He) | Fe56(H2) |
| Cr52(H2) | Ni58(He) |
| Fe54(He) | Ni58(H2) |
| Fe54(H2) | Ni60(He) |
| Fe56(He) | |

Table 28: Validated isotopes and gas conditions of major elements

| Validated minor elements | | | | | |
|--------------------------|----------|-----------|-----------|-----------|----------|
| Al27(H2) | Mn55(H2) | Mo95(He) | Sn118(He) | Pb206(He) | U238(H2) |
| Ti48(He) | Co59(H2) | Mo98(He) | Sn118(H2) | Pb208(He) | |
| Ti49(He) | Cu63(He) | Cd112(He) | W182(He) | Pb206(H2) | |
| V51(He) | Cu65(He) | Cd114(He) | W183(He) | Pb207(H2) | |
| V51(H2) | As75(He) | Cd112(H2) | W182(H2) | Pb208(H2) | |
| Mn55(He) | As75(H2) | Cd114(H2) | W183(H2) | U238(He) | |

Table 29: Validated isotopes and gas conditions of minor elements

Nevertheless, not all elements were considered as variables for the chemical profile. This is due to the fact that certain elements were not found or that the standard deviation of the 5 replicates was too elevated to be considered (see section 4.4.3.1).

4.4.3 ICP-MS results

The detected counts per seconds (cps) were transformed into parts per billion (ppb) according to the daily calibration. The obtained values were then corrected after considering the initial weight of the specimen cut and the dilution factor according to the following equation:

$$c_{final \text{ [ppb]}} = \frac{c_{solution}}{(w_{initial}/V_{am})} * 1000 * DF$$

where c_{final} is the final concentration, $c_{solution}$ the measured concentration in the solution, $w_{initial}$ the initial weight of the cut, V_{am} the final volume after the mineralization and DF the dilution factor.

4.4.3.1 CRI gas and isotope selection

Two different collision and reaction gases (He and H₂) were used to minimize possible polyatomic and isobaric interferences. Nonetheless, certain isotopes or gas conditions might be more reliable than others. A single isotope and gas condition per element were selected in a further step, which would ensure that each analysed element is represented by a single variable and thus prevent overweighing certain elements. The selection criteria were the following:

- The main criterion, in a profiling context, is homogeneity. Hence, variables with the lowest overall %RSD for all the measured replicates were favoured.
- If the %RSD was comparable, the preferred isotope was the one with the highest natural abundance, since it would be less likely to be subjected to errors of measurements than isotopes with a low natural abundance.

Table 30 and Table 31 show the values for the mean overall %RSD, natural abundance of the different isotopes and the retained variables that were considered for further data analysis.

Experimental section

| Element | Gas condition | Isotope | Mean overall RSD [%] | Natural abundance [%] | Selection |
|---------|----------------|---------|----------------------|-----------------------|------------------------------------|
| Cr | He | 52 | 4.48 | 83.79 | ^{52}Cr (He) |
| | H ₂ | 52 | 4.52 | | |
| Fe | He | 54 | 4.59 | 5.85 | ^{56}Fe (H ₂) |
| | H ₂ | 54 | 5.29 | | |
| | He | 56 | 4.56 | 91.75 | |
| | H ₂ | 56 | 4.43 | | |
| Ni | He | 58 | 4.39 | 68.08 | ^{58}Ni (He) |
| | H ₂ | 58 | 4.43 | | |
| | He | 60 | 4.39 | 26.22 | |

Table 30: All validated major elements, gas conditions and isotopes and selection of variables according based on mean overall %RSD of all replicates and natural abundance of isotopes

| Element | Gas condition | Isotope | Mean overall RSD [%] | Natural abundance [%] | Selection |
|---------|----------------|---------|----------------------|-----------------------|-------------------------------------|
| Al | H ₂ | 27 | 12.53 | 100 | ^{27}Al (He) |
| | He | 48 | 7.57 | 73.72 | |
| Ti | He | 49 | 10.19 | 5.41 | ^{48}Ti (He) |
| | H ₂ | 51 | 1.64 | 99.75 | |
| V | He | 51 | 1.93 | | ^{51}V (He) |
| | H ₂ | 51 | | | |
| Mn | He | 55 | 1.55 | 100 | ^{55}Mn (H ₂) |
| | H ₂ | 55 | 1.37 | | |
| Co | H ₂ | 59 | 1.74 | 100 | ^{59}Co (H ₂) |
| Cu | He | 63 | 3.37 | 69.17 | ^{63}Cu (He) |
| | He | 65 | 3.35 | 30.83 | |
| As | He | 75 | 5.97 | 100 | ^{75}As (He) |
| | H ₂ | 75 | 7.64 | | |
| Sn | He | 118 | 2.76 | 24.22 | ^{118}Sn (H ₂) |
| | H ₂ | 118 | 2.02 | | |
| W | He | 182 | 7.85 | 26.5 | ^{182}W (H ₂) |
| | H ₂ | 182 | 7.52 | | |
| | He | 183 | 7.76 | 14.31 | |
| | H ₂ | 183 | 7.44 | | |
| Pb | He | 206 | 15.59 | 24.1 | ^{208}Pb (H ₂) |
| | H ₂ | 206 | 3.87 | | |
| | H ₂ | 207 | 4.56 | 22.1 | |
| | He | 208 | 13.75 | 52.4 | |
| | H ₂ | 208 | 3.72 | | |

Table 31: All validated minor elements, gas conditions and isotopes and selection of variables according based on mean overall %RSD of all replicates and natural abundance of isotopes

4.4.3.2 Alloy identification

Steel is an alloy, which is a mixture of different metals, and Iron is its main constituent. All the elements that are intentionally added for a specific purpose (e.g. higher corrosion resistance) are called alloying elements [116].

Stainless steel is defined as steel containing more than 10% chromium. Chromium was found in all watchcases and the mean value of chromium is at 16.97 %. The advantages of stainless steel include corrosion resistance and the fact that it is easily maintained. It is widely used for a broad range of applications. Nickel is an alloying element, which increases corrosion resistance and offers better ability to reshape the alloy¹⁸. Our specimens are made of austenitic stainless steel, meaning that they are a chromium-nickel stainless steel class. Molybdenum was also found in all specimens and is also used to increase resistance to corrosion [116]. Even if Mo did not appear among the validated elements, it helped for the identification of the steel type.

A visual examination of the counterfeit watches of *specimen set 1* indicated that they were of relatively good quality. In addition, the entirety of the watches, including their mechanical components, were found to be in the higher quality segment of counterfeits. It should be noted that watches of poorer quality may be made of alloys other than steel.

4.4.3.3 Chemical composition of all specimens

In this section, only results for the validated and retained elements are shown. The values are presented as the mean value of the 5 measured replicates $\pm s_d$. Specimen *M01* was not measured using ICP-MS. XRF analysis revealed that *M01* was made of a Zn-Cu-Ni alloy and the dissolution matrix was exclusively developed for steel. The results are summarized in Table 32, Table 33 and Table 34, in which the major elements are given in g·kg⁻¹ and the minor elements in ppb¹⁹.

¹⁸ Information retrieved from the Steel Glossary of the American Iron and Steel Institute website: <http://www.steel.org/steel-technology/glossary/glossary-a-e.aspx> (last accessed: 02.03.2020)

¹⁹ 1 ppb = 0.000001 g·kg⁻¹ = 0.001 mg·kg⁻¹

Experimental section

| Watch | ^{52}Cr (He) [g/kg] | ^{56}Fe (H ₂) [g/kg] | ^{58}Ni (He) [g/kg] |
|-------|------------------------------|---|------------------------------|
| M02 | 157.03±2.9 | 659.61±12.26 | 97.12±1.41 |
| M03 | 172.55±1.33 | 699.69±9.05 | 77.59±0.91 |
| M04 | 174.08±2.19 | 702.59±7.75 | 80±1.17 |
| M05 | 170.27±1.48 | 700.45±5.31 | 78.89±0.36 |
| M06 | 167.04±3.59 | 678.43±6.35 | 77.35±2.38 |
| M07 | 165.71±0.37 | 686.46±4.82 | 77.92±0.46 |
| M08 | 169.06±2.05 | 690.76±1.86 | 78.52±0.55 |
| M09 | 170.43±1.09 | 673.25±6.24 | 77.86±0.7 |
| M10 | 173.56±3.01 | 670.02±13.41 | 77.97±1.37 |
| M11 | 169.97±1.71 | 672.12±8.59 | 78.42±2.3 |
| M12 | 170.85±1.79 | 666.62±6.05 | 78.22±0.75 |
| M13 | 168.78±2.86 | 674.61±14.03 | 78.62±1.77 |
| M14 | 166.62±3.96 | 665.15±18.79 | 77.13±2.36 |
| M15 | 168.8±9.79 | 665.22±39.58 | 75.93±4.41 |
| M16 | 176.06±3.01 | 692.02±12.99 | 79.95±2.38 |
| M17 | 174.8±9.34 | 683.18±36.7 | 80.23±3.94 |
| M18 | 169.67±2.82 | 677.08±14.38 | 74.95±1.19 |
| M19 | 169.18±4.31 | 671.2±13.59 | 78.8±1.34 |
| M20 | 173.33±0.93 | 691.59±3.51 | 77.16±0.82 |
| M21 | 170.14±1.52 | 656.41±8.64 | 71.92±1.32 |
| M22 | 170.8±2.91 | 672.24±9.47 | 77.25±2.31 |
| M23 | 166.28±3.38 | 681.6±11.13 | 79.68±1.85 |
| M24 | 187.76±1.51 | 689.48±6.18 | 85.12±0.58 |
| M25 | 179.99±3.37 | 684.04±13.1 | 80.49±1.41 |
| M26 | 177.45±11.87 | 668.01±44.85 | 80.6±3.61 |
| M27 | 187.85±3.01 | 701.8±11.51 | 84.82±1.28 |
| M28 | 185.19±5.6 | 683.73±24.05 | 79.7±2.05 |
| M29 | 185.81±3.71 | 695.56±14.21 | 82.05±1.63 |
| M30 | 159.49±5.61 | 603.79±24.14 | 70.4±1.78 |
| M31 | 170.98±3.52 | 674.95±13.83 | 85.99±0.73 |
| M32 | 179.27±4.27 | 696.09±12.71 | 79.67±0.95 |
| M33 | 171.29±3.17 | 689.01±8.17 | 77.87±0.81 |
| M34 | 171.16±3.84 | 682.3±11.53 | 82.12±2.19 |
| M35 | 164.24±7.33 | 665.02±25.37 | 79.33±1.33 |

Table 32: ICP-MS results for major elements for the 34 specimens in [g·kg⁻¹]

Experimental section

| Watch | $^{27}\text{Al} (\text{H}_2)$ | $^{48}\text{Ti} (\text{He})$ | $^{51}\text{V} (\text{He})$ | $^{55}\text{Mn} (\text{H}_2)$ | $^{59}\text{Co} (\text{H}_2)$ | $^{63}\text{Cu} (\text{He})$ | $^{75}\text{As} (\text{He})$ | $^{118}\text{Sn} (\text{H}_2)$ | $^{182}\text{W} (\text{H}_2)$ | $^{208}\text{Pb} (\text{H}_2)$ |
|-------|-------------------------------|------------------------------|-----------------------------|-------------------------------|-------------------------------|------------------------------|------------------------------|--------------------------------|-------------------------------|--------------------------------|
| M02 | 9.43±1.24 | 17.61±0.5 | 857.97±11.28 | 11676.31±344.29 | 2081±73.72 | 583.17±31.08 | 21.14±1.4 | 15.61±0.46 | 82.67±4.32 | 0.25±0.01 |
| M03 | 4.82±0.85 | 4.47±1.1 | 898.32±21.49 | 13697.26±237.32 | 2670.61±31.67 | 609.89±21.55 | 23.15±1.87 | 31.45±0.54 | 16.57±4.1 | 0.4±0.01 |
| M04 | 7.77±3.4 | 27.52±2.2 | 865.37±13.7 | 12505.14±184.51 | 2270.38±47.18 | 181.36±13.27 | 18.16±1.25 | 7.96±0.18 | 21.81±2.26 | 0.26±0.01 |
| M05 | 24.96±0.91 | 46.24±0.92 | 1101.58±7.06 | 8925.7±120.19 | 2214.61±39.13 | 403.48±7.71 | 23.03±0.83 | 10.6±0.21 | 14.15±0.44 | 0.56±0.02 |
| M06 | 33.7±0.46 | 21.82±0.69 | 721.98±2.38 | 12490.53±114.24 | 1912.27±21.82 | 1290.95±2.77 | 33.28±0.72 | 32.81±0.41 | 35.03±0.11 | 0.93±0 |
| M07 | 25.33±0.67 | 4.71±0.01 | 755.37±1.72 | 12608.2±15.73 | 2097.39±17.62 | 1013.99±11.17 | 27.27±1.69 | 36.62±0.42 | 22.09±0.32 | 2.75±0.03 |
| M08 | 4.32±0.72 | 13.4±2.49 | 507.12±12.16 | 12487.05±142.62 | 2257.81±31.1 | 194.71±7.19 | 18.42±0.57 | 6.09±0.1 | 17.81±2.39 | 0.55±0.02 |
| M09 | 35.53±4.05 | 8.74±1.2 | 897.29±8.76 | 19370.12±151.11 | 2078.25±23.89 | 403.08±17.88 | 20.61±1.69 | 12.92±0.13 | 26.62±1.91 | 0.61±0.02 |
| M10 | 28.98±7.71 | 9.21±0.87 | 891.69±10.54 | 20242.73±342.24 | 2001.16±26.44 | 363.92±13.75 | 19.12±1.54 | 11.69±0.13 | 21.96±2.32 | 0.64±0.02 |
| M11 | 28.92±6.33 | 9.17±0.57 | 897.6±6.66 | 20151.35±144.54 | 1985.83±25.88 | 368.2±10.64 | 17.79±1.2 | 11.59±0.19 | 21.44±2 | 0.62±0.01 |
| M12 | 39.65±7.19 | 7.71±1.35 | 846.44±16.29 | 19982.99±156.27 | 2111.09±28.21 | 413.66±11.56 | 20.89±1.46 | 13.14±0.14 | 26.34±4.03 | 0.67±0.02 |
| M13 | 7.32±0.43 | 30.91±1.58 | 905.39±12.47 | 13226.71±121.46 | 2176.2±37.67 | 234.95±6.22 | 36.93±1.95 | 10.5±0.16 | 34.05±4.97 | 0.18±0.01 |
| M14 | 7.29±0.28 | 32.07±0.83 | 927.63±17.91 | 13262.77±71.56 | 2193.33±31.24 | 235.19±3.8 | 36.28±0.76 | 10.51±0.18 | 36.6±0.27 | 0.16±0.01 |
| M15 | 6.87±0.26 | 32.36±0.84 | 931.36±4.92 | 13198.65±62.48 | 2180.97±27.64 | 236.17±10.86 | 36.53±2.1 | 10.51±0.16 | 36.54±0.28 | 0.14±0.01 |
| M16 | 4.56±0.81 | 19.66±2.79 | 1023.96±16.6 | 13156.57±193.99 | 1949.83±18.75 | 269.96±4.7 | 16.87±1.03 | 10.48±0.07 | 25.95±3.95 | 0.28±0.01 |
| M17 | 5.82±1.45 | 26.02±3.36 | 1106.58±82.95 | 14212.59±840.13 | 2377.48±122.44 | 235.62±9.22 | 12.48±1.08 | 9.27±0.63 | 23.13±5.08 | 0.29±0.02 |
| M18 | 26.52±2.04 | 4.28±0.4 | 963.48±12.7 | 12724.29±155.91 | 1870.18±21.19 | 1948.82±54.94 | 31.25±1.83 | 42.49±0.8 | 45.16±1.25 | 0.86±0.01 |

Table 33: ICP-MS results for minor elements (M02-M18) in [ppb]

Experimental section

| Watch | ^{27}Al (H ₂) | ^{48}Ti (He) | ^{51}V (He) | ^{55}Mn (H ₂) | ^{59}Co (H ₂) | ^{63}Cu (He) | ^{75}As (He) | ^{118}Sn (H ₂) | ^{182}W (H ₂) | ^{208}Pb (H ₂) |
|-------|------------------------------------|-----------------------|----------------------|------------------------------------|------------------------------------|-----------------------|-----------------------|-------------------------------------|------------------------------------|-------------------------------------|
| M19 | 52.32±6.05 | 14.93±2.15 | 975.89±12.29 | 15076.15±90.11 | 2082.31±13.09 | 331.72±7.82 | 17.82±1.31 | 12.34±0.23 | 28.02±3.51 | 0.29±0.01 |
| M20 | 6.69±0.78 | 32.29±1.43 | 975.71±3.37 | 13232.34±26.92 | 1897.18±20.23 | 226.75±11.67 | 13.1±0.95 | 5.97±0.06 | 21.15±1.59 | 0.64±0.01 |
| M21 | 30.97±2.37 | 14.89±0.74 | 972.44±3.82 | 19714.93±64.18 | 2049.75±57.2 | 351.19±2.08 | 18.62±1.45 | 10.08±0.22 | 21.04±0.33 | 0.54±0.01 |
| M22 | 33.34±1.46 | 7.15±0.59 | 753.37±10.86 | 16647.59±68.3 | 2192.19±31.59 | 358.55±13.14 | 19.66±1.26 | 12.96±0.16 | 39.6±4.14 | 0.74±0.01 |
| M23 | 29.88±2.2 | 14.96±3.39 | 791.18±16.8 | 9203.8±112.56 | 2154.6±38.28 | 359.37±7.31 | 18.31±1.77 | 12.44±0.17 | 31.03±4.71 | 0.64±0.01 |
| M24 | 16.5±0.87 | 20.39±0.78 | 883.79±11.22 | 11251.03±62.07 | 1958.18±17.28 | 753.82±16.49 | 22.52±0.89 | 17.36±0.14 | 39.12±0.17 | 2.95±0.05 |
| M25 | 7.82±0.66 | 31.07±0.4 | 866.55±1.64 | 12795.14±35.57 | 2085.83±14.86 | 217.55±6.61 | 35.3±1.34 | 10.1±0.13 | 35.41±0.58 | 0.18±0.01 |
| M26 | 11.06±3.74 | 25.98±1.25 | 939.91±13.01 | 12867.05±102.34 | 2015.83±10.73 | 268.17±16.85 | 21.86±2.14 | 4.74±0.15 | 8.96±0.37 | 0.64±0.05 |
| M27 | 8.58±1.26 | 27.98±1.34 | 983±21.44 | 13526.17±330.76 | 2071.5±56 | 268.1±17.07 | 24.33±1.28 | 5.02±0.23 | 8.95±0.5 | 0.75±0.04 |
| M28 | 6.49±0.32 | 17.31±0.43 | 1347.77±13.53 | 10075.7±245.43 | 1977.34±79.27 | 745.4±44.8 | 21.16±0.98 | 9.84±0.25 | 8.46±0.39 | 0.17±0.02 |
| M29 | 6.83±2.39 | 27.82±0.61 | 988.29±11.73 | 12747.33±78.76 | 2241.49±40.85 | 193.52±4.86 | 20.91±0.9 | 18.32±0.15 | 11.52±0.23 | 0.45±0.01 |
| M30 | 36.47±3.51 | 10.64±0.85 | 937.55±31.32 | 21214.75±607.91 | 2033.72±65.58 | 263.54±9.94 | 18.68±1.61 | 8.79±0.24 | 20.74±1.16 | 0.66±0.03 |
| M31 | 23.04±0.7 | 9.04±0.3 | 746.26±3.44 | 17236.31±254.15 | 2357.98±28.87 | 299.92±9.38 | 20.04±0.86 | 8.1±0.12 | 39.99±0.99 | 1.76±0.02 |
| M32 | 4.81±0.37 | 27.92±0.77 | 1131.03±17.82 | 14379.47±261.36 | 2086.45±18.25 | 266.88±7.05 | 18.39±1.44 | 10.88±0.26 | 29.22±0.35 | 0.3±0.01 |
| M33 | 42.77±3.73 | 30.01±1.28 | 1045.53±6.53 | 13778.05±167.27 | 2206.36±25.72 | 629.49±18.85 | 22.96±0.57 | 16.63±0.19 | 17.37±2.22 | 0.37±0.01 |
| M34 | 24.29±0.63 | 17.49±0.48 | 794.77±18.35 | 13115.28±255.63 | 2192.41±51.19 | 904.4±46.31 | 42.37±1.03 | 30.63±0.74 | 64.49±0.81 | 0.58±0.01 |
| M35 | 41.01±3.61 | 37.06±3.41 | 950.44±62.42 | 15584.91±520.66 | 2118.77±66.79 | 608.59±15.53 | 22.76±1.54 | 16.87±1.04 | 29.73±2.88 | 0.46±0.05 |

Table 34: ICP-MS results for minor elements (M19-M35) in [ppb]

4.5 Portable XRF analysis on *specimen set 1*

Portable X-ray fluorescence analysis (pXRF) was performed on all specimens using an Oxford X-MET7500 handheld pXRF instrument with a 45kV Rh target X-ray tube and a silicon-drift detector. The instrument was used with a benchtop stand, this setup is recommended to analyse small specimens and to protect the user from scattered radiation. Nevertheless, it is possible to do the same measurements in the field without using the benchtop stand.



Picture 1: Oxford X-MET7500 handheld XRF instrument with benchtop stand

4.5.1 Analytical procedure

No specimen preparation was performed, since the goal of the measurements were to investigate the possibility of obtaining useful information without altering the watch in any manner (by disassembling for example). The watchcases were only cleaned with 70% EtOH prior to measurement.

The 35 watchcases of *specimen set 1* were each analysed at five different points on the surface of the cases. For *specimen set 2*, the cases and backs were analysed for all 35 watches, the straps and clasps were analysed only if the strap was made of metal. Three measurements at different locations, including three replicate measurements were performed on each watch part. All specimens of *specimen set 2* that indicated a higher than 10% Ni presence during pXRF analysis were tested with a colorimetric method using test strips (110006, Nickel Test, Merck). This method is a simple way to identify nickel in alloys or nickel-plated surfaces like jewelry. The surface was wiped with a test

strip soaked in a 10% solution of ammonia. The formation of a pink dye indicates the release of nickel. There was no quantification of the nickel content, but the results indicated a substantial release.

4.5.2 Quality controls for pXRF (*specimen set 1* and *specimen set 2*)

Quality controls were carried out to demonstrate that the pXRF procedure is suitable for the intended application and included:

- Analysis of blank samples (silicone dioxide)
- The accuracy was tested using quality control samples consisting of certified reference materials. The certified reference materials were chosen with respect to the alloy compositions of the specimen set, as explained below.
- Precision was assessed through repeated analysis of the same specimens
- Certified reference materials. For the analysis, different certified reference materials were chosen to cover all elements found during the ICP-MS analysis for *specimen set 1*, as well as legally regulated elements for *specimen set 2*.
 - Low-alloy steel (MBH Analytical)
 - Copper-Nickel alloy (MBH Analytical)
 - Nickel Waspalloy (MBH Analytical)
 - Paint chips (CRM13-50G, Sigma-Aldrich)

4.5.2.1 Blank samples

The blank matrix consisted of silicon dioxide (SiO_2 Matrix Blank, Sigma-Aldrich) in the form of sand, free of any analytes of interest. The blanks were analysed at the beginning of each working day, as well as after every fifth specimen. For both, *specimen set 1* and *specimen set 2*, none of the elements found during the analysis of blank samples were observed. Hence the method blank can be considered acceptable [127].

4.5.2.2 Accuracy and precision

The ‘accuracy’ and ‘precision’ relate to the same definitions as given in section 4.4.2.2. The recovery and repeatability were tested for several elements in *specimen set 1* and shown in Table 35 and Table 36. Only elements that were systematically found in each specimen were considered. In order to assess repeatability, the %RSD values should

not exceed 15%. Since the samples of *specimen set 1* were intended to be used in a chemical profiling context, specific elements that did not fulfil the repeatability requirements were not selected. Also, elements that were not systematically present, like Ti and Cu, Zn, Mo, Ag and W in *specimen set 1* could not be considered, since these elements were present in relatively low concentrations. The risk of measurement errors is higher in this case and/or these elements are not homogeneously distributed within the alloy and therefore not systematically detected.

| CRM | Element | Certified conc. [% by weight] | Mean measured conc. [% by weight] | Recovery [%] | SD [ppb] | %RSD | # Meas. |
|------------------------|---------|----------------------------------|--------------------------------------|--------------|----------|-------|------------|
| Low-alloy steel | Cr | 2.1 | 2.413 | 114.89 | 0.032 | 1.33 | 60 |
| | Ni | 0.4 | 0.325 | 81.23 | 0.017 | 5.22 | 60 |
| | Mn | 1.1 | 1.234 | 112.14 | 0.030 | 2.46 | 60 |
| | Co | 0.55 | 0.624 | 113.43 | 0.032 | 5.13 | 60 |
| Copper-Nickel alloy | Fe | 2.63 | 3.081 | 117.16 | 0.045 | 1.48 | 60 |
| | Cr | 0.097 | 0.109 | 112.70 | 0.028 | 25.31 | 60 |
| | Ni | 65.3 | 65.323 | 100.03 | 0.123 | 0.19 | 60 |
| | Mn | 1.105 | 1.199 | 108.48 | 0.028 | 2.32 | 60 |
| Nickel Waspalloy | Fe | 0.997 | 1.064 | 106.68 | 0.034 | 3.22 | 60 |
| | Cr | 19.52 | 20.740 | 106.25 | 0.136 | 0.65 | 60 |
| | Ni | 57.2 | 57.122 | 99.86 | 0.170 | 0.30 | 60 |
| | Co | 13.31 | 13.490 | 101.35 | 0.099 | 0.74 | 60 |

Table 35: Recovery percentages between the measured (recovered) and certified value and statistical parameters for the elements measured by pXRF

Cr showed a very high %RSD in the Copper-Nickel alloy CRM. However, Cr showed satisfying values in the Low-alloy steel CRM, as well as in the Nickel-Waspalloy CRM. These two CRMs found Cr in a higher concentration (2.1% and 19.52%). Since Cr is present in high concentrations (\approx 18% by weight) in the watch cases, we decided not to exclude Cr based on these findings. All recovery values lay within the acceptable range of 80% - 120% of the measured CRM concentration.

From *specimen set 1*, the specimens M15 and M20 were randomly chosen to assess repeatability of the measurement, they were each measured 21 times (three measurements before and after every fifth specimen).

Experimental section

| Specimen | Element | Mean [g/kg] | SD [g/kg] | %RSD | Selected |
|----------|---------|-------------|-----------|-------|----------|
| M15 | Cr | 18.58 | 0.09 | 0.48 | ✓ |
| | Mn | 1.39 | 0.06 | 4.08 | ✓ |
| | Fe | 71.73 | 0.16 | 0.22 | ✓ |
| | Co | 0.27 | 0.04 | 16.41 | ✗ |
| | Ni | 7.87 | 0.09 | 1.10 | ✓ |
| M20 | Cr | 18.55 | 0.11 | 0.59 | ✓ |
| | Mn | 1.35 | 0.06 | 4.09 | ✓ |
| | Fe | 71.88 | 0.20 | 0.28 | ✓ |
| | Co | 0.23 | 0.04 | 16.65 | ✗ |
| | Ni | 7.85 | 0.10 | 1.33 | ✓ |

Table 36: Repeatability values for specimens M15 and M20 of *specimen set 1* analysed with pXRF

In both specimens, Co showed a %RSD higher than 15% and was excluded as a potential profiling variable.

4.5.3 pXRF results for *specimen set 1*

The elements that were found are Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Mo, Ag and W. The main constituents were Cr, Fe and Ni. The elements that were found in all 34²⁰ specimens were Cr, Mn, Fe, Co and Ni. Except for Co, these elements were validated and constitute potential profiling variables. The software uses a fundamental parameters method to calculate the concentration of each element and the data is constrained to 100% for each analysis.

²⁰ Specimen M01 was not analysed using pXRF since its alloy composition was found to be fundamentally different from the other 34 specimens.

Experimental section

| Watch | Cr [%] | Mn [%] | Fe [%] | Ni [%] |
|-------|------------|-----------|------------|------------|
| M02 | 17.28±0.10 | 1.21±0.06 | 69.23±0.08 | 10.08±0.07 |
| M03 | 18.46±0.08 | 1.54±0.05 | 71.18±0.17 | 7.74±0.09 |
| M04 | 18.62±0.10 | 1.38±0.07 | 71.67±0.12 | 7.94±0.12 |
| M05 | 18.32±0.11 | 1.01±0.05 | 72.19±0.18 | 7.97±0.15 |
| M06 | 18.09±0.09 | 1.33±0.03 | 72.41±0.17 | 7.75±0.07 |
| M07 | 18.13±0.11 | 1.4±0.10 | 72.09±0.07 | 7.94±0.08 |
| M08 | 18.42±0.10 | 1.42±0.05 | 71.89±0.10 | 7.9±0.05 |
| M09 | 18.58±0.05 | 2.11±0.05 | 70.56±0.06 | 8.06±0.06 |
| M10 | 18.58±0.09 | 2.21±0.06 | 70.64±0.15 | 7.99±0.06 |
| M11 | 18.71±0.06 | 2.22±0.09 | 70.48±0.10 | 8.01±0.06 |
| M12 | 18.6±0.10 | 2.12±0.07 | 70.56±0.08 | 8.02±0.06 |
| M13 | 18.6±0.17 | 1.38±0.10 | 71.67±0.19 | 7.91±0.17 |
| M14 | 18.49±0.11 | 1.45±0.09 | 71.74±0.09 | 7.94±0.11 |
| M15 | 18.57±0.09 | 1.45±0.05 | 71.64±0.12 | 7.95±0.11 |
| M16 | 18.52±0.14 | 1.43±0.05 | 71.7±0.23 | 7.98±0.10 |
| M17 | 18.49±0.12 | 1.39±0.10 | 71.73±0.12 | 7.95±0.09 |
| M18 | 18.02±0.06 | 1.25±0.08 | 72.78±0.20 | 7.49±0.11 |
| M19 | 18.05±0.06 | 1.58±0.06 | 71.75±0.08 | 8.1±0.10 |
| M20 | 18.55±0.05 | 1.35±0.04 | 71.87±0.12 | 7.8±0.08 |
| M21 | 18.66±0.09 | 2.04±0.08 | 70.97±0.09 | 7.88±0.10 |
| M22 | 18.32±0.13 | 1.82±0.03 | 71.12±0.12 | 8.1±0.10 |
| M23 | 18.04±0.04 | 1.02±0.05 | 71.36±0.10 | 8.26±0.07 |
| M24 | 18.66±0.09 | 1.22±0.03 | 70.49±0.12 | 8.01±0.06 |
| M25 | 18.62±0.07 | 1.46±0.04 | 70.89±0.07 | 7.58±0.05 |
| M26 | 18.7±0.07 | 1.47±0.06 | 70.78±0.05 | 7.82±0.02 |
| M27 | 18.8±0.06 | 1.41±0.04 | 70.71±0.06 | 7.77±0.04 |
| M28 | 18.84±0.11 | 1.08±0.03 | 71.21±0.14 | 7.55±0.08 |
| M29 | 18.58±0.13 | 1.36±0.02 | 71.14±0.12 | 7.65±0.12 |
| M30 | 18.79±0.13 | 2.22±0.06 | 69.98±0.18 | 7.62±0.06 |
| M31 | 18.05±0.12 | 1.79±0.02 | 69.57±0.08 | 8.36±0.06 |
| M32 | 18.65±0.09 | 1.42±0.05 | 70.71±0.11 | 7.74±0.10 |
| M33 | 18.25±0.07 | 1.43±0.03 | 70.99±0.10 | 7.63±0.11 |
| M34 | 18.12±0.14 | 1.26±0.07 | 71.2±0.07 | 8.12±0.07 |
| M35 | 17.76±0.09 | 1.56±0.10 | 71.81±0.26 | 7.94±0.13 |

Table 37: pXRF results of validated elements for the 34 specimens [% by weight]

4.6 Portable XRF analysis on *specimen set 2*

4.6.1 pXRF results for *specimen set 2*

Only results that are relevant in terms of potentially toxic heavy metals are presented here. Table 38 shows the results for the tested watch parts that are very close or exceed (in bold) the legal limit for lead in Switzerland and Europe (0.05% by weight).

| Specimen | Location | Concentration of Pb [%] | | | |
|----------|----------|-------------------------|---------------------|---------------------|---------------------|
| | | Measure 1 | Measure 2 | Measure 3 | Measure 4 |
| M29 | Case | 0.058 ± 0.009 | 0.65 ± 0.019 | 1.10 ± 0.020 | 1.04 ± 0.002 |
| M31 | Case | 0.05 ± 0.003 | 0.05 ± 0.008 | ND ²¹ | 0.03 ± 0.006 |
| M31 | Strap | 1.73 ± 0.022 | 2.13 ± 0.024 | 1.86 ± 0.006 | 2.21 ± 0.026 |
| M32 | Case | 0.93 ± 0.014 | 0.91 ± 0.026 | 0.98 ± 0.020 | 0.92 ± 0.008 |

Table 38: Concentration of lead found in specimen M29, M31 and M32

Table 39 shows the results of high lead concentration found on 2 specimens decorated with imitated gemstones.

| Specimen | Concentration of Pb [%] | | | |
|-------------------------------------|-------------------------|----------------------|----------------------|----------------------|
| | Measure 1 | Measure 2 | Measure 3 | Measure 4 |
| M20 (bezel and strap) ²² | 14.23 ± 0.106 | 21.49 ± 0.095 | ND | ND |
| M23 (bezel) | 26.12 ± 0.073 | 21.81 ± 0.071 | 22.19 ± 0.040 | 26.48 ± 0.014 |

Table 39: Concentration of lead found on the imitated gemstones on specimen M20 and M23

4.6.2 Nickel release test

Table 40 shows the results from the test strips. The formation of a pink dye indicates the presence of released nickel. Where a stronger pink colour indicates that a higher amount of nickel was released onto the test strip. The obtained measurement results are only indicative.

²¹ ND: not detected

²² Measure 1 and 2 were performed on the bezel, whereas measure 3 and 4 were performed on the strap.

Experimental section

| Watch | Part | Result | Photo |
|-------|-------|-------------------|-------|
| M20 | Clasp | Negative | |
| M24 | Back | Negative | |
| M26 | Case | Positive | |
| M30 | Strap | Slightly positive | |
| M31 | Back | Positive | |
| M31 | Case | Slightly positive | |
| M31 | Clasp | Positive | |
| M32 | Strap | Negative | |
| M34 | Case | Positive | |
| M34 | Strap | Positive | |

Table 40: Watch parts that were tested for potential nickel release and corresponding results from the presumptive colorimetric test

4.7 Method comparison (Portable XRF vs. ICP-MS)

From an analytical point of view, it is interesting to compare the techniques that were chosen to measure the elemental concentration.

Experimental section

First of all, it is important to mention that pXRF is a surface analysis technique. The surface of the objects may not be representative of their bulk composition, which was measured by ICP-MS. Interpreting the differences between the pXRF and ICP-MS results may be complex [128].

A striking difference between the two methods can be seen in the number of elements that were measured. Table 41 summarises the elements that were systematically detected in all specimens, and the ones that were validated and can be used as profiling variables.

| | pXRF | ICP-MS |
|--------------------------------|--------------------|--|
| Systematically detected | Cr, Mn, Fe, Co, Ni | Cr, Fe, Ni, Al, Ti, V, Mn, Co, Cu, Zn, As, Mo, Sn, W, Pb |
| Validated | Cr, Mn, Fe, Ni | Cr, Fe, Ni, Al, Ti, V, Mn, Co, Cu, As, Mo, Sn, W, Pb |

Table 41: Elements detected and validated for both, pXRF and ICP-MS

Another factor that needed to be considered was that the pXRF software sums all the selected elements to 100%. To compare pXRF results with those of the ICP-MS, only Cr, Fe and Ni were selected in the pXRF. The data was standardized to 100% for the three elements. All the replicate measurements were used. Correlation between the data obtained from the two techniques is shown in Figure 12.

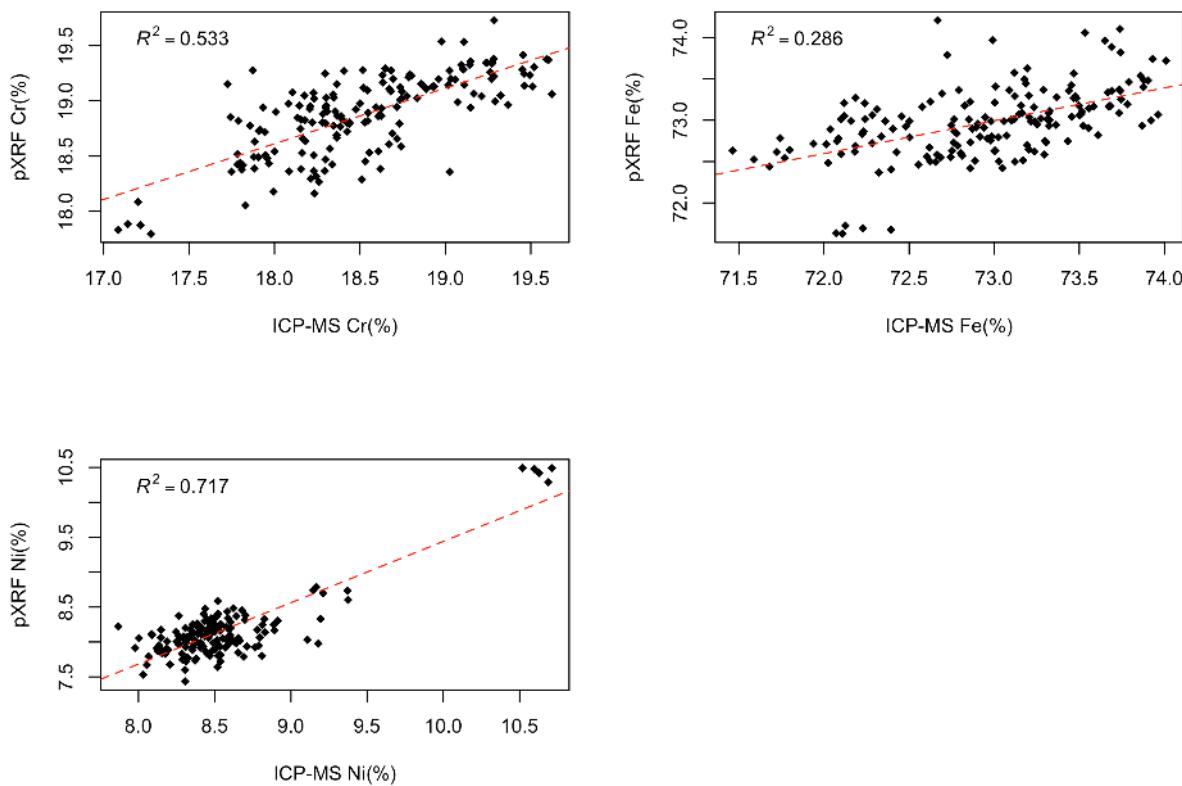


Figure 12: Linear relationship between the measurements of major elements (Cr, Fe and Ni) by XRF and ICP-MS

The determination coefficients are low, especially for Cr and Fe, indicating a weak linear relationship between the two measurements. Furthermore, a paired *t*-test was performed to compare the concentrations obtained by ICP-MS and pXRF (Table 42).

| | Paired Differences | | | | <i>t</i> | <i>df</i> | <i>p</i> | | | |
|--------------------|--------------------|-------|---|--------|----------|-----------|----------|--|--|--|
| | Mean | SD | 95% Confidence interval of the difference | | | | | | | |
| | | | Lower | Upper | | | | | | |
| Cr(ICP) – Cr(pXRF) | 0.564 | 0.405 | 0.422 | 0.704 | 8.107 | 33 | .000 | | | |
| Fe(ICP) – Fe(pXRF) | -0.804 | 0.315 | -0.913 | -0.694 | -14.897 | 33 | .000 | | | |
| Ni(ICP) – Ni(pXRF) | 0.24 | 0.224 | 0.162 | 0.318 | 6.258 | 33 | .000 | | | |

Table 42: ICP-pXRF paired *t*-test results

The element by element paired *t*-tests show significant differences for all elements between the ICP-MS and pXRF determined concentrations ($p < 0.001$). Both, correlation and hypothesis testing, showed that for the individual elements the reading concentrations are significantly different. Nevertheless, aggregating the measured

Experimental section

elements into chemical profiles could provide a comparable grouping of the specimens into chemically resembling classes. This issue will be examined more closely in the next chapter.

5 Chemical profiling

The aim of this chapter was to propose a statistical methodology to discriminate between watchcases having common and different origins based on their measured chemical resemblance. All the watchcases from *specimen set 1* were analysed by ICP-MS and pXRF. For both analytical techniques, several elements were validated and used for profiling. Several issues were raised, and possible solutions were proposed.

This section has been divided into three parts: The first and the second part focus on chemical profiling using ICP-MS and pXRF, respectively. In the third part, a comparison of the grouping using the two different data sources was performed.

Data processing was performed using Microsoft Excel 2016 and R (version 3.4.2) in combination with RStudio (version 1.1.383).

5.1 ICP-MS

5.1.1 Major elements

The major elements that were found in the 34 watchcases are Cr, Fe and Ni. The distribution of the 34 specimens considering these three major elements is visualised in a 3D- Scatterplot (see Figure 13).

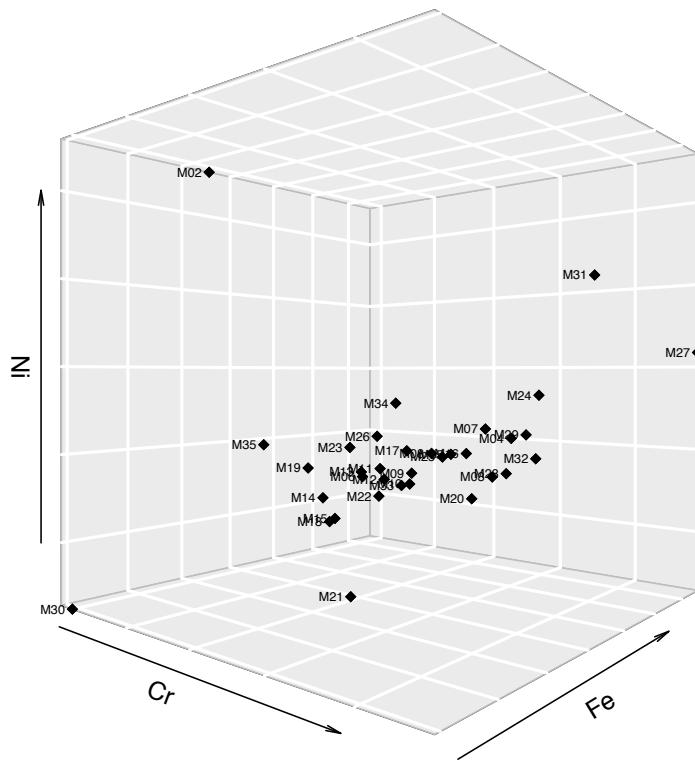


Figure 13: 3D Scatterplot of the Cr, Fe and Ni detected in the 34 specimens

Some specimens clearly differ from the others, based on their Ni (M02, M31 and M27) or Fe (M30 and M21) content. The vast majority of the specimens are joined at the centre with no discernible structure. It is conceivable that the main composition of the steel alloys is quite similar between watchcases coming from different production lines. Thus, better linking and/or separation between the specimens might be achieved through exploitation of the trace element profiles. In addition, the laborious analysis required for ICP-MS would not justify its use for determining the major elemental composition. Furthermore, ICP-MS is a technique intended for quantitative ultra-trace element and isotope analysis [122].

Combining major and minor elements would also not be very reasonable due to their extreme differences in magnitude. Hence, statistically comparing them would render the minor elements relatively insignificant or an extreme transformation of the data would be required to obtain a comparable scale. The latter, however, might disproportionately influence errors of measurements, especially for variables with minor contributions. For this reason, the major elemental compositions measured by ICP-MS were not used and the minor elements were focused on for chemical profiling.

5.1.2 Minor elements

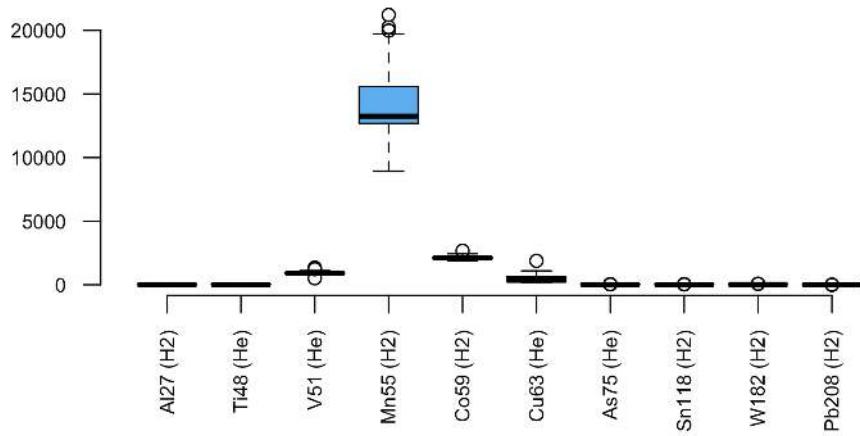
This section considers the chemical profiling of the trace metal composition (minor elements) from the ICP-MS results. Several multivariate approaches were assessed, considering four scenarios, each including a different set of variables. The objective of this process was to evaluate the effect of variable selection to construct the profile on the subsequent multivariate data treatment.

5.1.2.1 *Data structure and variable selection*

Before exploratory data analysis, it is advantageous to examine the data structure and prepare the data. A simple and informative way of representing the distribution of each variable is using boxplots, which are based on the median and quantiles of the data. Raw-data exploration revealed large differences in the magnitude of trace metals, which necessitated data pre-processing to achieve better symmetry. It should be noted, that the analytical information has to be maintained and care should be taken to avoid amplifying background noise; and there is a risk of disproportionately influencing errors of measurements, particularly for values that are close to their LOQ.

Amongst the multitude of available pre-processing techniques, after testing, it was decided to scale the data using a cube root transformation. Power transformations, where x_{ij} is replaced by $(x_{ij})^{1/n}$, reduce the influence of large values. The distribution of the unprocessed raw data and cube root transformed data for all potential variables can be found in Figure 14.

Raw Data



CRT

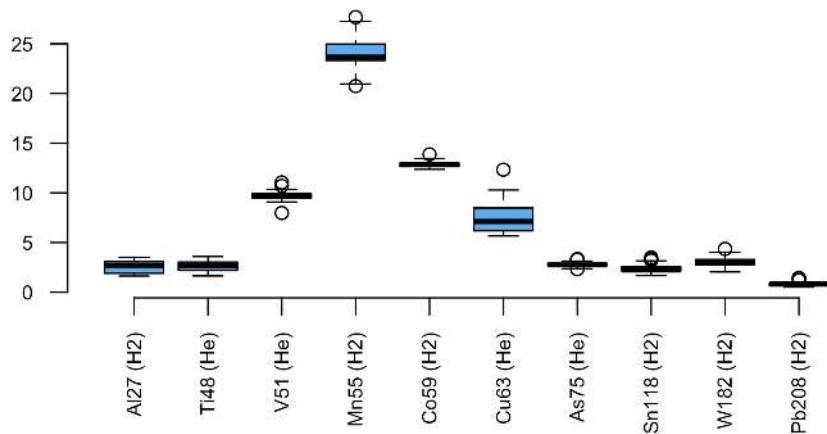


Figure 14: Representation of the variability of all retained variables in the raw data and on pre-processed data after cube root transformation (CRT)

5.1.2.2 Inter- and intravariability

Since the goal of chemical profiling is to link watchcases from a common source and to discriminate between watchcases from different sources, inter- and intra-source variations needed to be measured. The counterfeit watches that we had access to were seized by custom authorities and no formal indication of a common source existed. A '*common source*' refers to a situation where two watchcases were produced in the same facility with the same raw material (e.g. the same steel basis) and machinery. Therefore, the intra-source variation had to be determined by other means. For this, two options

were considered. The first option was to consider the variation of the five sample cuts from a single specimen. In the absence of specimens known to originate from a common source, measuring the specimen-intravariability is the only possibility to know whether the two measurements are linked. Considering specimen-intravariability means that we assume the variability within a specimen reflects the variability between specimens of a common source. Nevertheless, the ‘true’ variation between different specimens of a common source might be greater than the determined specimen-intravariability.

Another option was then to consider watches that were seized together. Several watches that were seized at the same time and place also showed high similarity in terms visual examination, technical features (e.g. movement and functionalities) and physical profiling. For the physical profiling²³, characteristics such as the tagging of brand names, logos and symbols present on the watch were not distinguishable on any part of the watch (case, back, dial and strap). It can be assumed that these specific seizures possibly emanated from a common production route source. We refer to this as being the seizure-intravariability. Six groups with 14 watches were complied with the aforementioned requirements and are shown in Figure 15. The intervariability was determined by taking into consideration watchcases from unrelated seizures.

²³ Physical profiling of counterfeit watches is explained in detail in Chapter 6

| Group | Watches | Image |
|----------------|--------------------|-------|
| Group 1 | M09, M10, M11, M12 | |
| Group 2 | M13, M25 | |
| Group 3 | M14, M15 | |
| Group 4 | M26, M27 | |
| Group 5 | M28, M29 | |
| Group 6 | M30, M31 | |

Figure 15: Groups of counterfeit watches (with corresponding images) belonging to the same seizures and being indistinguishable based on visual examination as well as on technical and physical characteristics

5.1.2.3 Scenarios

In the next step, four different scenarios, comprising combinations of the selected variables, were then evaluated. The different scenarios included the following combinations of the selected variables:

- **Scenario 1:** The profile includes the complete set of variables
Al, Ti, V, Mn, Co, Cu, As, Sn, W, Pb

- **Scenario 2:** The variables for scenario 2 were those measured at the highest concentration ($> 100 \text{ ppb}$), since they are the most likely to be remeasured again in future steel case samples

V, Mn, Co, Cu

- **Scenario 3:** For scenario 3, the retained variables were those whose concentration were in a comparable analytical range ($10 \text{ ppb} < x < 100 \text{ ppb}$), to reduce the influence of very high or very low concentrations

Al, Ti, As, Sn, W

- **Scenario 4:** Variables with the lowest overall %RSD were used, because these elements were potentially the most homogeneously distributed elements within the watchcases

V, Mn, Co, Cu, As, Sn

Intravariability distributions were computed by measuring the Euclidean distance between sample pairs from a specific specimen (specimen-intravariability), and between sample pairs from different watches but the same seizure group (seizure-intravariability). Intervariability was computed between the replicate pairs from watches of unrelated seizures. During this process, pairs between a specific specimen, as well as pairs from different watches but from the same seizure group were excluded. It is, however, important to mention that an unrelated seizure context does not mean that two specimens could not be linked. For both seizure intravariability and intervariability distributions, we considered the mean value of five samples per specimen as well as all the five samples, for the four aforementioned combinations of variables (scenarios 1 to 4). The obtained distributions, including the number of comparisons in each category, are presented in Figure 16.

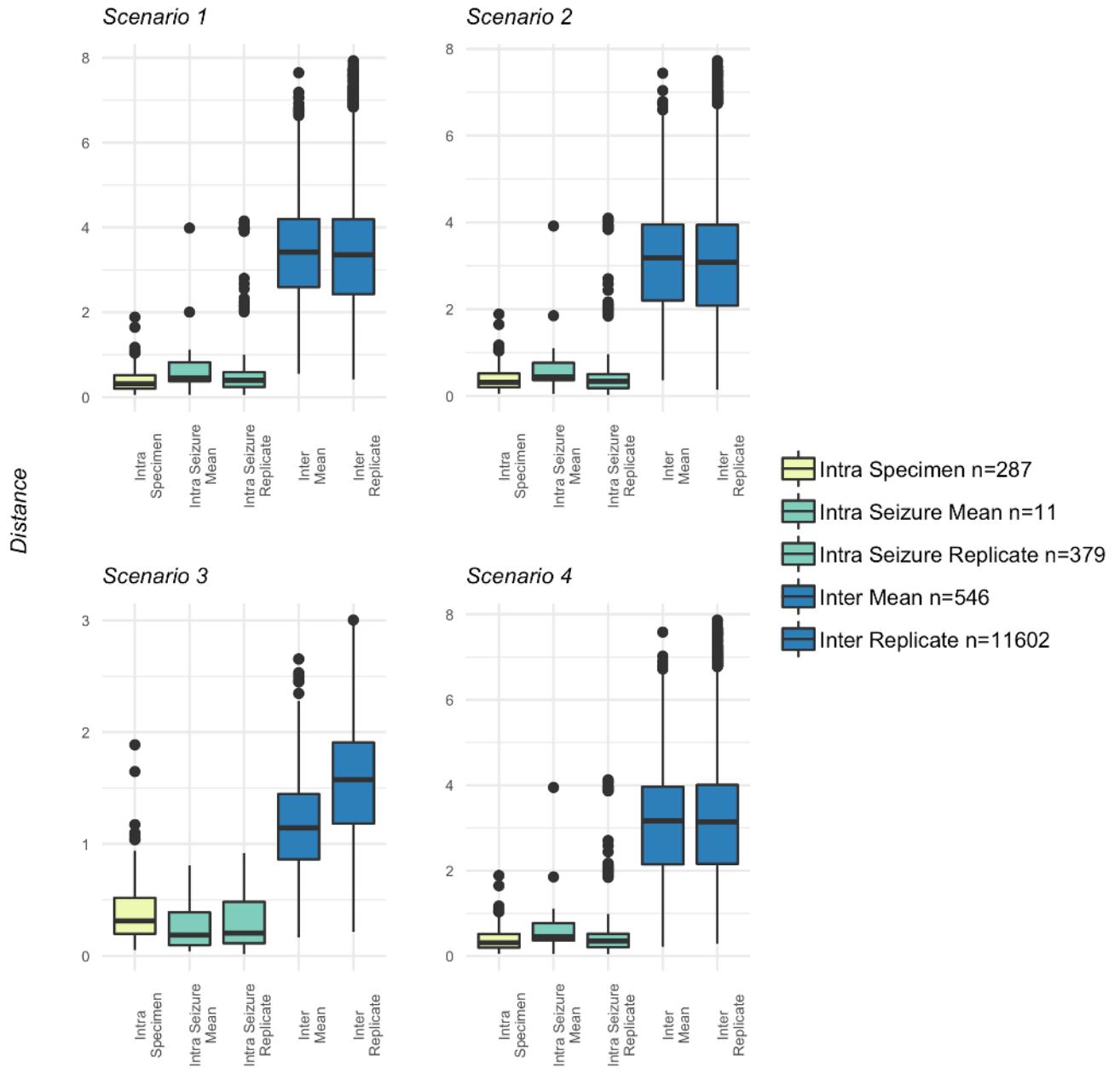


Figure 16: Boxplots representing the distributions of specimen-intravariability, seizure-intravariability (mean and replicates) and intervariability (mean and replicates)

In all four scenarios we observe that, on the one hand, both the specimen-intravariability and seizure-intravariability are low, while on the other hand, the intervariability computed between specimens from unrelated seizures is high. This suggests a separation between the intra- and inter-source variations, and at the same time the possibility to distinguish watchcases from common and different production sources, based on measurements of their elemental composition. As can be seen from the above figure, scenarios 1, 2 and 4 exhibit a robust behaviour when considering the

mean values or all the replicate samples, although the number of comparisons in the latter is much larger. Scenario 3 displayed a higher sensitivity to such changes. An explanation for this could be that the considered elements are less homogeneously distributed and/or present in very low concentration compared to some other elements. Consequently, there is also a higher risk of measurement errors. The same observations can also be made when representing the specimen-intravariability and intervariability for the different scenarios in a density plot (see Figure 17). The results of the pairwise distance comparisons between samples of the same specimen (specimen-intravariability) are represented by the green curve, while for watchcases belong to unrelated seizures (intervariability), the distribution takes the shape of the red curve. In all four scenarios the red curve covers a broader range of distance values and is more dispersed and flattened than the green curve. This indicates that the comparison between sample cuts from the same specimen produces distances which are small.

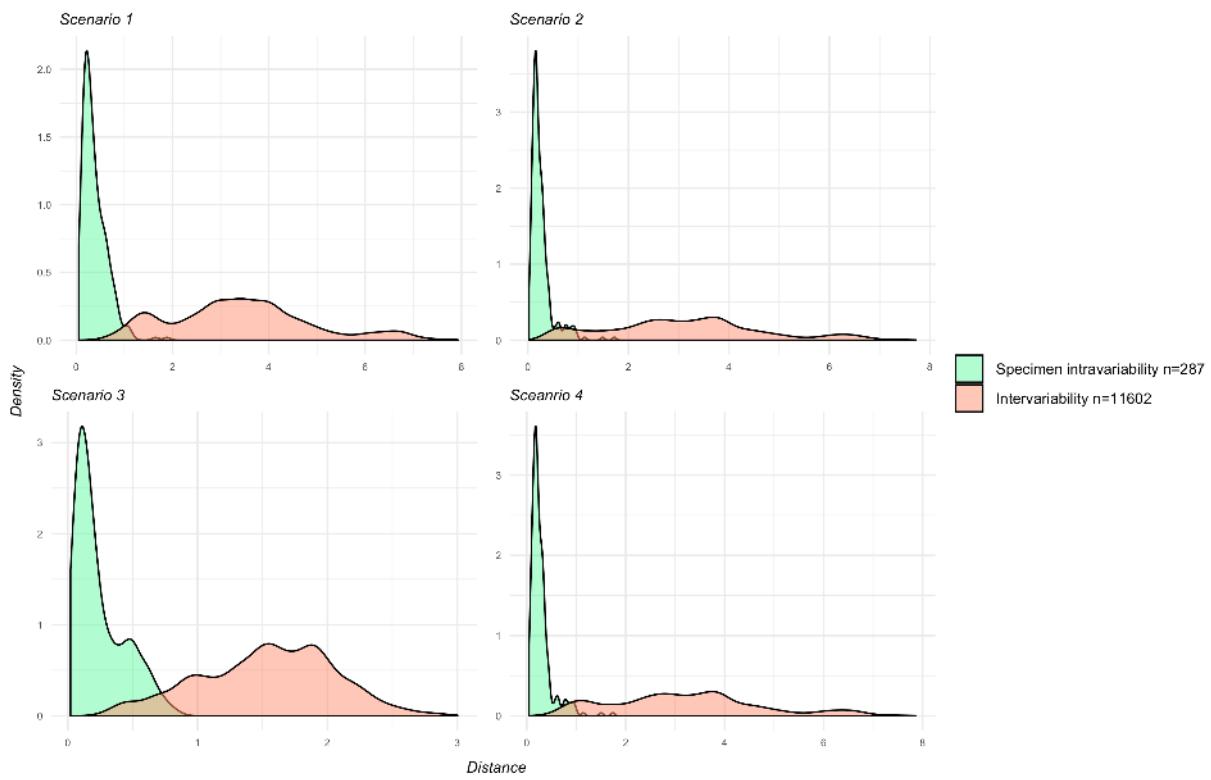


Figure 17: Density plots showing the distribution of specimen-intravariability and intervariability for all four scenarios

5.1.2.4 Model evaluation

In order to infer a common source, the question at stake then, is to define what 'close enough' means. Linking or discriminating watchcases based on their chemical profile, required the definition of a threshold value. As mentioned earlier in section 3.4.2, in an intelligence-based perspective we attempt to maximise the true positive (TP) rate. If a link does exist, we do not want to exclude it from further evaluation, even if a certain number of false positives (FP) may also be included. The specimen-intravariability is an approximation for the unknown 'true' production-intravariability. We assume that the production-intravariability is greater, or at any rate, equal to the specimen-intravariability. On these grounds, the strategy to set the threshold values for each scenario at the maximum distance computed for the specimen-intravariability was adopted.

The performance of each scenario in discriminating the seizure-intravariability from the intervariability was then evaluated. The sensitivity (true positive rate) and specificity (true negative rate) at the given threshold (maximum distance computed for specimen-intravariability) were calculated (Figure 18). Also, receiver operating characteristic (ROC) curves were plotted and the corresponding area under the curve (AUC) was computed (Figure 19). All relevant metrics for all scenarios are summarized in Table 43.

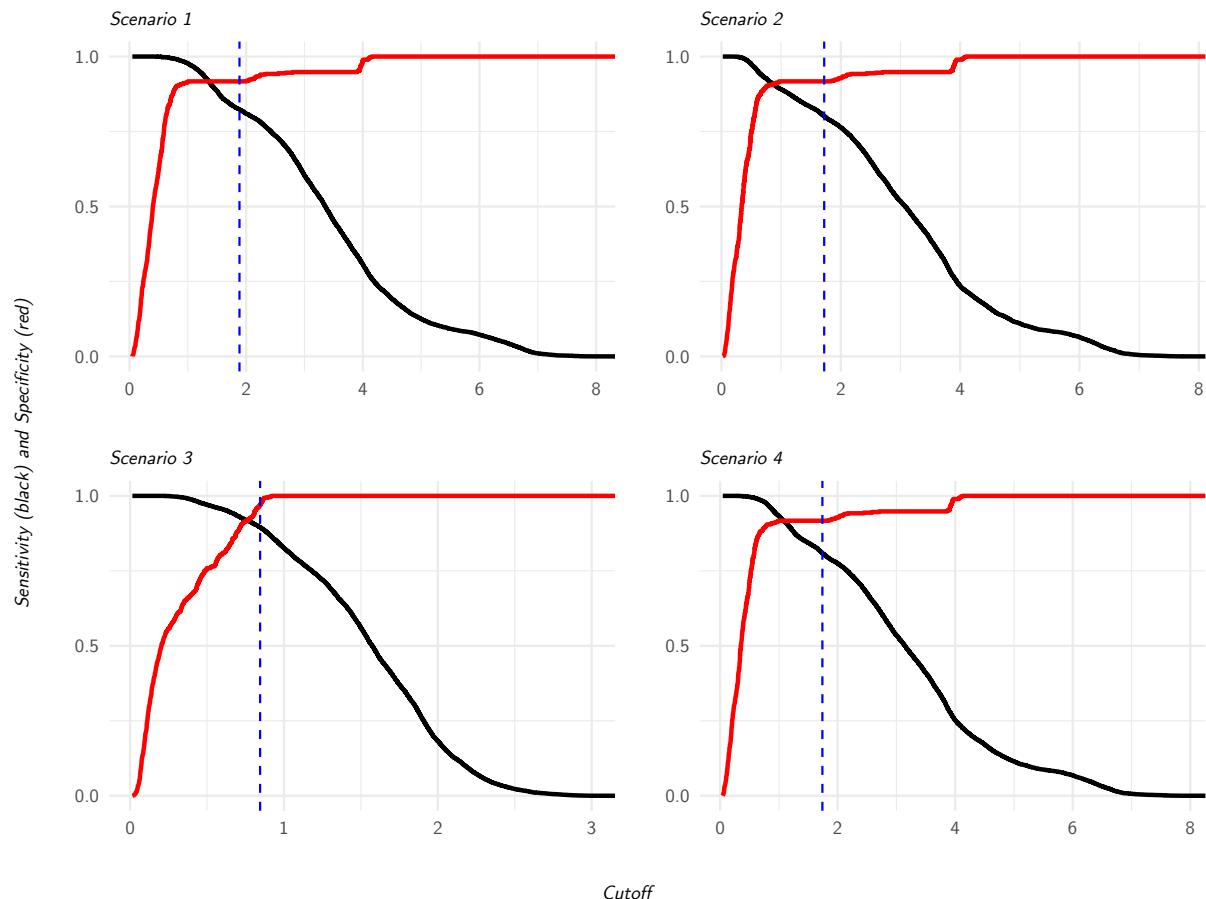


Figure 18: Sensitivity (true positive rate) in black and specificity (true negative rate) in red as a function of the cut-off distance. The dashed blue line represents the threshold value that was set at the highest specimen-intravariability value for each scenario.

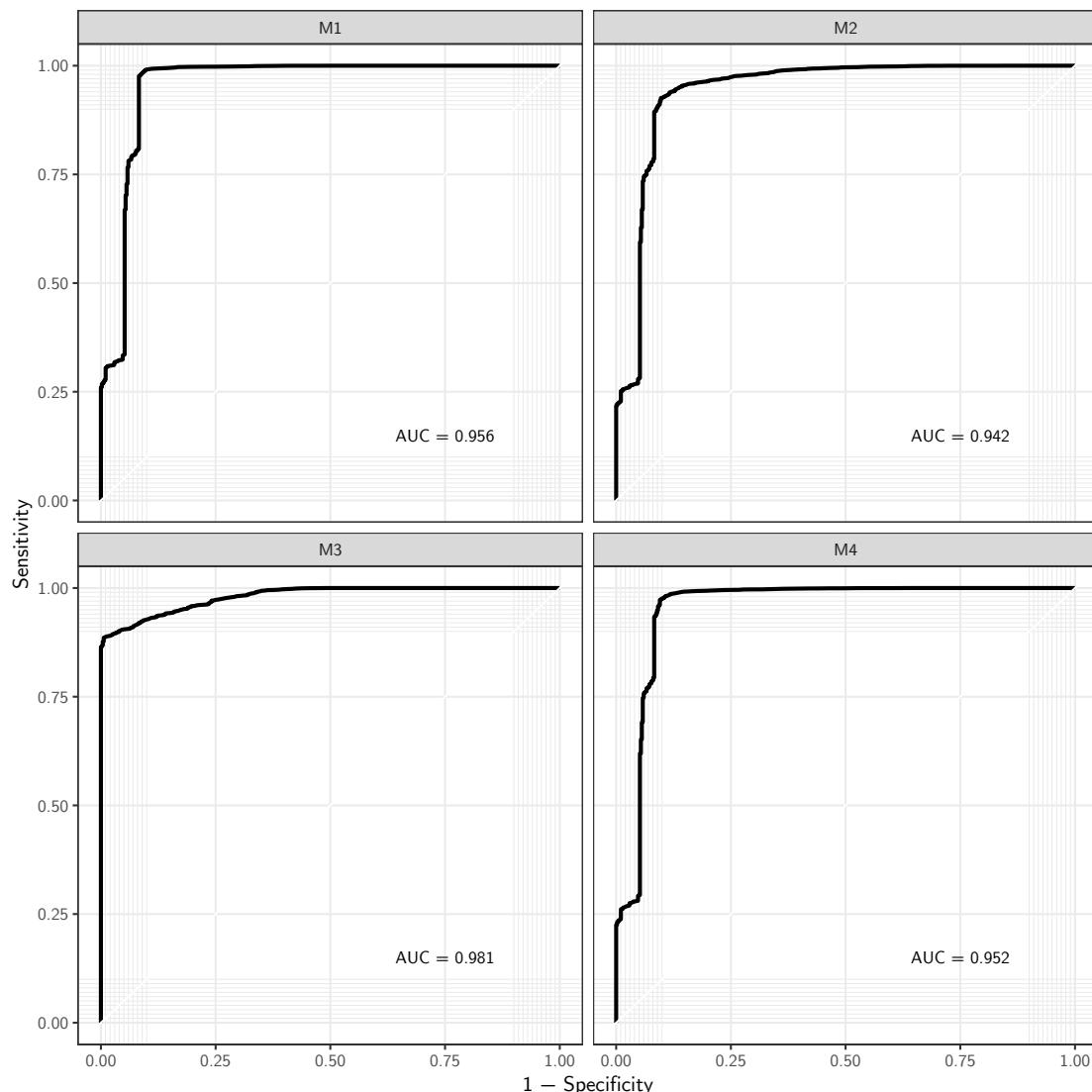


Figure 19: ROC curves for each scenario including the respective AUC values

| | Threshold | Sensitivity | Specificity | AUC |
|-------------|-----------|-------------|-------------|-------|
| Scenario 1: | 1.886 | 0.917 | 0.823 | 0.956 |
| Scenario 2: | 1.723 | 0.917 | 0.801 | 0.942 |
| Scenario 3: | 0.845 | 0.969 | 0.895 | 0.981 |
| Scenario 4: | 1.74 | 0.917 | 0.809 | 0.952 |

Table 43: Performance criteria for all scenarios, including sensitivity and specificity at the given threshold and AUC values for the ROC curves

On a first look at the results, scenario 3 appears to be the model that performs the best. For this scenario, five variables were included over a comparable analytical range ($10 \text{ ppb} < x < 100 \text{ ppb}$). At the set threshold distance, the sensitivity, specificity and

AUC were higher than for the three other scenarios. Nevertheless, the AUC values for all the scenarios were high (>0.94), indicating a reliable classification of new specimens of the watchcases. The sensitivity was exactly the same for scenarios 1, 2 and 4, but the specificity and AUC were all also very similar and seem to produce the same outcome.

Hence, further consideration was given to the distribution of the seizure-intravariability and its comparison with the specimen-intravariability, as well as the intervariability, as shown in Figure 20.

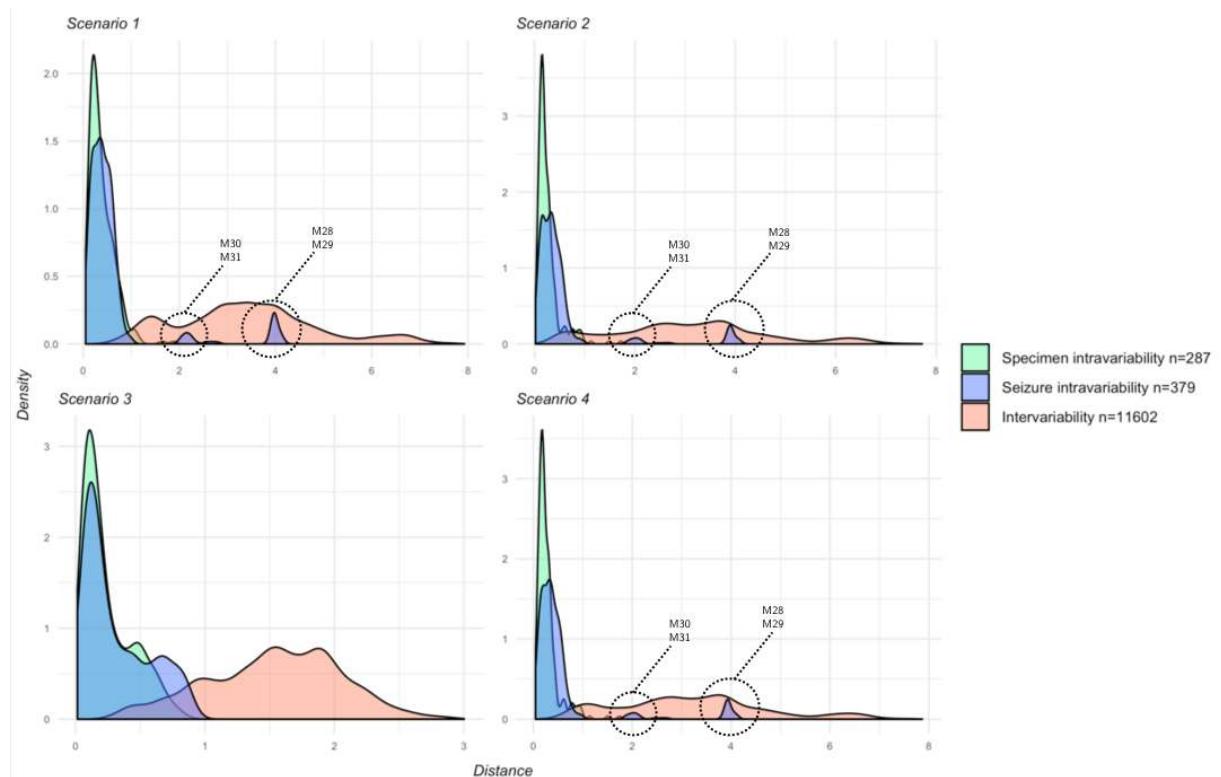


Figure 20: Density plots showing the distribution of specimen-intravariability, seizure-intravariability and intervariability for all four scenarios. Distance measurement values lying outside the specimen-intravariability are encircled with a dotted line. From Hochholdinger et al. [129].

Generally, there is good agreement between the computed specimen- and seizure-intravariability (green and blue curve, respectively). Selected watches that were visually similar and had the same seizure context, also showed high similarity in their chemical profile. Scenarios 1, 2 and 4 have the common characteristic that they include the elements Mn, Co, and Cu into the profile, these are also among the most abundant minor elements. This also means that these elements contribute the most to the distance measurements, consequently the distribution curves of the three scenarios also exhibit similar shapes. Nevertheless, two small peaks at distances of approximately 2 and 4 appeared in the seizure-intravariability distribution. By investigating the distance matrices of the 6 seizure groups, it was found that these peaks correspond to the watch pairs M28/M29 and M30/M31. These peaks are discernibly within the intervariability distribution, suggesting that the two watch pairs should not be linked. In stark contrast, the watch pairs in scenario 3 are included amongst the specimen-intravariability. We can observe that the specimen- and seizure-intravariability are almost superimposable. Some distance measurements found in the seizure group M09/M10/M11/M12 have a few values that are a little higher than the specimen-intravariability. However, in scenario 1, 2 and 4, this particular seizure group is completely absorbed within the specimen-intravariability. For all the above reasons, scenario 3 would not be an appropriate choice even if its classification performance seems superior. The elements Mn, Cu and Co are abundant and could potentially contribute greatly to the distance measurements. In addition, they showed very low overall %RSD, which indicates a homogeneous distribution within the alloy and makes them important profiling variables.

The differences in the variable selection between the three remaining scenarios had very little influence on the performance criteria. However, scenario 1 appeared to have a slightly better specificity and AUC. It was decided to choose the scenario 1 for further evaluation, including all ten variables and use the distance 1.886 as a threshold value for clustering. Based on this, two watchcases separated by a distance less than the threshold value were supposed to be chemically linked and the inference that they were produced from the same source is supported.

The next two sections explore the relationships between the specimens and between the variables through PCA, followed by dividing the specimens into classes, with similar specimens in the same class by HCA.

5.1.2.5 Principal component analysis (PCA)

A specimen grouping tendency was evaluated using PCA. The visualisation of the plot for the first and second principal component can be found in Figure 21. Different colours were assigned for the specimens according to their watch model. The specimen distribution does not allow identifying distinct groups.

For a specific model given, it is never the case that all specimens are close to each other. But some specimens of a particular model can be found very closely, e.g. M09/M10/M11/M12/M21. On the other hand, the group in the top centre is composed of three different models (Cosmograph Daytona, Datejust and Day-Date).

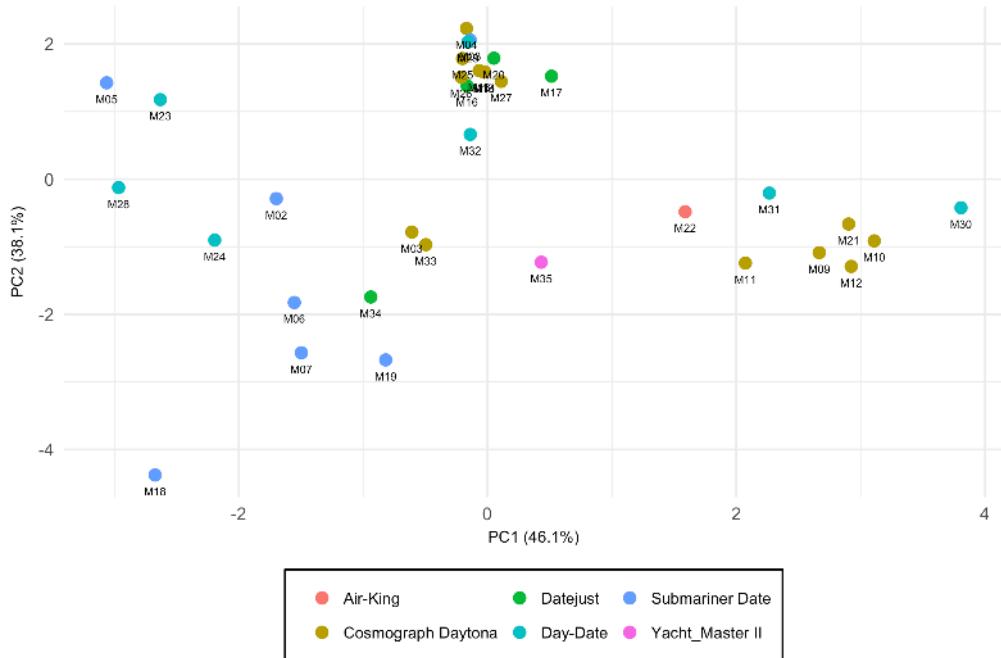


Figure 21: Repartition of the watchcases as a function of the first (PC1) and the second (PC2) principal component. The first two principal components express 84.2% of the total variance. The colors indicate the different watch models shown in the legend.

The contribution of each variable (in scenario 1) was visualised for dimensionality reduction. PC1 is the first dimension (new ‘x-axis’) and PC2 the second one (new ‘y-axis’), as well as a scree plot showing how much variance is explained by each of the

principal components (see Figure 22). In our case, the first two principal components explain 84.2% of the total variance in the data.

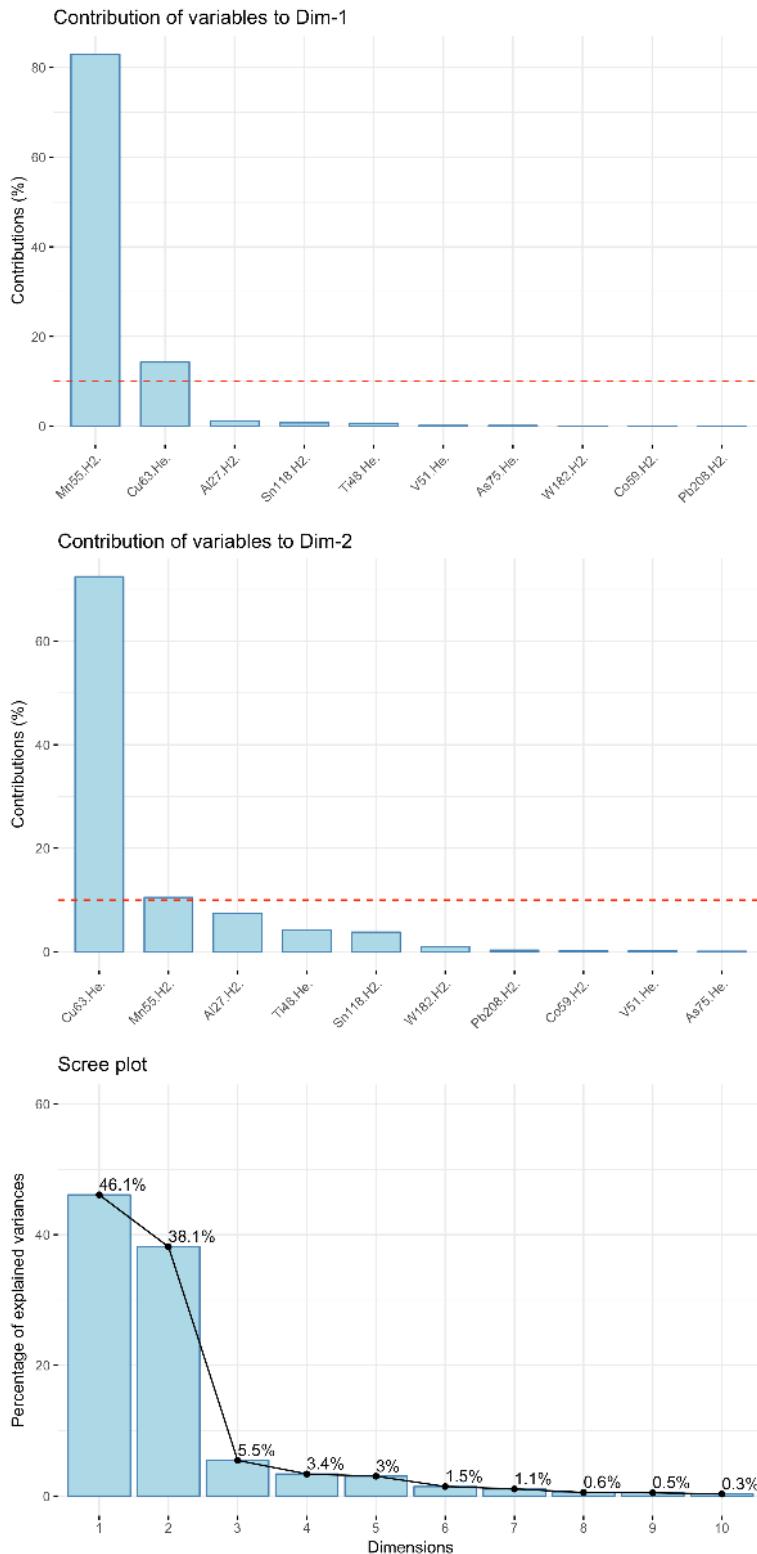


Figure 22: Bar plots of variable contribution to PC1 (on top) and PC2 (in the middle) and scree plot representing the percentage of explained variance of each principal component (on the bottom)

The red dotted reference line in the bar plots correspond to the expected value if the contributions were uniform. A variable with a contribution above this reference line contributes significantly to the component. In particular, it can be seen that Mn and Cu contribute the most to both dimensions. This finding also justifies the choice of scenario 1 instead of scenario 3, in the latter these two elements were not included in the profile.

Co and V rank among the most abundant elements, but they account only for a very small proportion. The remaining elements also did not contribute significantly to either PC1 or PC2.

It is likely that scenarios 2 and 4 (also including Mn and Cu) would have produced similar results, which is supported by the fact that we observed similar intra- and intervariability distributions and similar values for the performance of the binary classification (linked vs. not lined).

Although the PCA may reveal groups of similar objects, it is not always successful in doing so. Cluster analysis, on the other hand, is specifically intended to search for groups.

5.1.2.6 Hierarchical cluster analysis (HCA)

In hierarchical cluster analysis, the splitting or merging clusters is based on their similarity, i.e. distance between the clusters. We used the Euclidean distance to measure the similarity between two specimens, which was already used to assess intra- and intervariability. Other distance measurements may also be considered. The mean value obtained for all 5 replicates per specimen was used. The applied agglomeration method was the Ward's method and the obtained dendrogram is shown in Figure 23. The previously determined threshold value is indicated by the black dashed line and allowed for the construction of 14 groups. The different groups are associated with different colours and are now defined as distinct chemical classes. Table 44 includes information relative to the watches in each group.

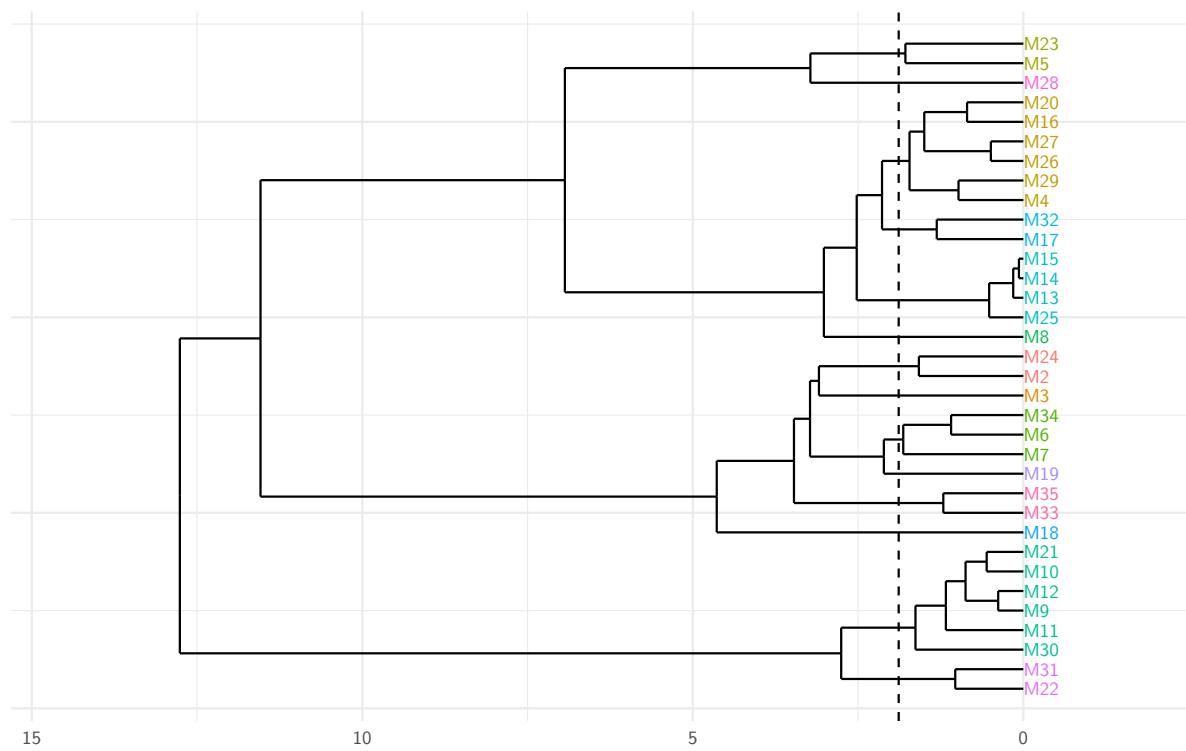


Figure 23: Dendrogram showing the results of the HCA, the black dashed line indicates the threshold that was used to form the 14 groups. From Hochholdinger et al. [129]

Chemical profiling

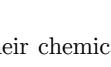
| Group | Watch/ Model | Picture | Group | Watch/ Model | Picture |
|-------|----------------|---|-------|------------------|---|
| 1 | M23 Day-Date |  | 8 | M03 Daytona |  |
| | M05 Submariner |  | | M34 Datejust |  |
| 2 | M28 Day-Date |  | 9 | M06 Submariner |  |
| | M20 Datejust |  | | M07 Submariner |  |
| 3 | M16 Datejust |  | 10 | M19 Submariner |  |
| | M27 Daytona |  | 11 | M35 Yacht-Master |  |
| 4 | M26 Daytona |  | | M33 Daytona |  |
| | M29 Day-Date |  | 12 | M18 Submariner |  |
| 5 | M04 Daytona |  | | M21 Daytona |  |
| | M32 Day-Date |  | | M10 Daytona |  |
| 6 | M17 Datejust |  | 13 | M12 Daytona |  |
| | M15 Datejust |  | | M09 Daytona |  |
| 7 | M14 Datejust |  | | M11 Daytona |  |
| | M13 Daytona |  | | M30 Day-Date |  |
| 8 | M25 Daytona |  | 14 | M31 Day-Date |  |
| | M08 Submariner |  | | M22 Airking |  |
| 9 | M24 Day-Date |  | 15 | M01 Deepsea |  |
| | M02 Submariner |  | | | |

Table 44: Watch specimens according to their chemical classes obtained through HCA using the ICP-MS results, including the watch model of each specimen and image of the watchcase

5.2 pXRF

The primary focus of the chemical profiling was directed by the ICP-MS results, nevertheless, the pXRF results are also worth investigating. Despite it being shown that the two analytical methods do not agree (see section 4.7), useful initial subdivision of the watchcases into the main chemical classes may also be achieved using pXRF results.

For analysis using the pXRF data, a similar methodology as for the ICP-MS results was used. An inspection of the data structure was undertaken first, as a next step, the intra- and inter-source variations were measured. The performance for the binary classification system was evaluated and a threshold value was assigned. Following this, the data structure was examined for patterns using PCA; and finally, a grouping was performed using HCA. Due to the detailed description previously provided, each step in this section is only briefly described.

5.2.1 Data structure

It was found that the four elements measured by pXRF showed only a very small variance for the 34²⁴ watchcases (see Table 45 and Figure 24). Since little variability was observed and the measured elemental concentration is in a comparable range, no pre-processing techniques were applied.

| | Cr | Mn | Fe | Ni |
|----------------|-------|-------|-------|------|
| Mean | 18.41 | 1.52 | 71.20 | 7.96 |
| S _d | 0.35 | 0.34 | 0.78 | 0.43 |
| %RSD | 1.88 | 22.47 | 1.10 | 5.38 |

Table 45: Mean, standard deviation and relative standard deviation for the 4 elements measured by pXRF

²⁴ Result for M01 was again discarded from evaluation due to a different alloy composition.

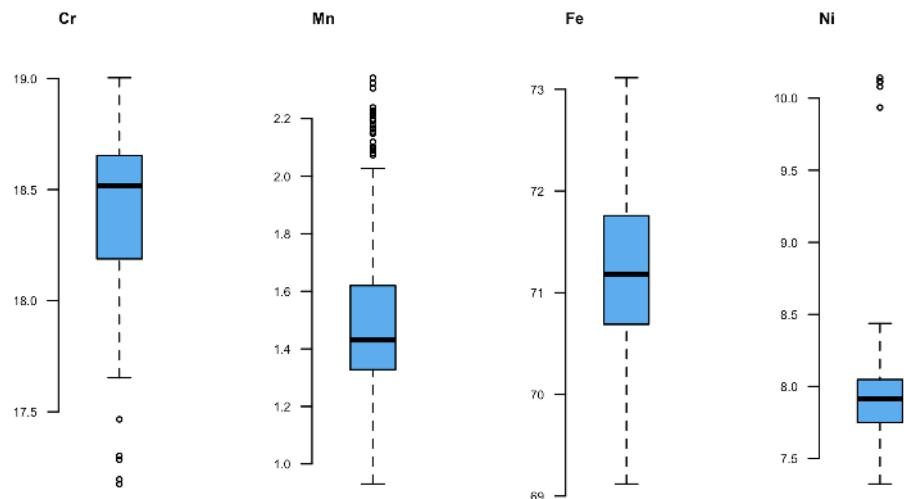


Figure 24: Representation of the variability of the measured concentration of the four elements measured by pXRF

5.2.2 Inter – and intravariability

Specimen intravariability, seizure intravariability and intervariability distributions were again computed by measuring the Euclidean distance. For *seizure intravariability* and *intervariability* distributions, the mean value of the five measurements per specimen as well as all five measurements were considered.

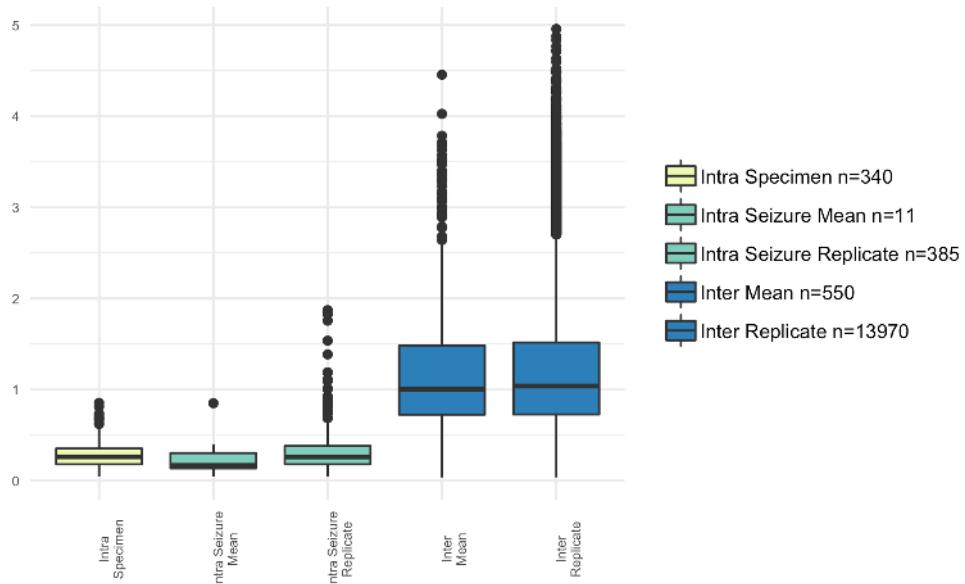


Figure 25: Boxplots representing the distributions of specimen-intravariability, seizure-intravariability (mean and replicates) and intervariability (mean and replicates) for pXRF measurements

As shown in Figure 25, the specimen- and seizure intravariability are low, whereas the intervariability is high. This finding suggests a possible separation of the watchcases based on their surface composition. In the present case, choosing the mean or replicate measurements had little influence, even if the number of comparisons between them was very different.

5.2.3 Model evaluation

Specimen intravariability and intervariability distributions are visualized by a green and red curve in the density plot, respectively. The same figure also includes the density plot with the seizure-intravariability distribution in blue (see Figure 26). The intervariability distribution, which covers a broad range values, is dispersed and much more flattened than the specimen-intravariability curve. Again, within the seizure intravariability (corresponding to a comparison between the watches M13/M25), some values exceed the specimen intravariability. Nevertheless, the model indicates the possibility to distinguish between the two populations that were defined as intra- and inter-source variations, which is also reinforced by the high AUC value (0.955) of the ROC curve (see Figure 27).

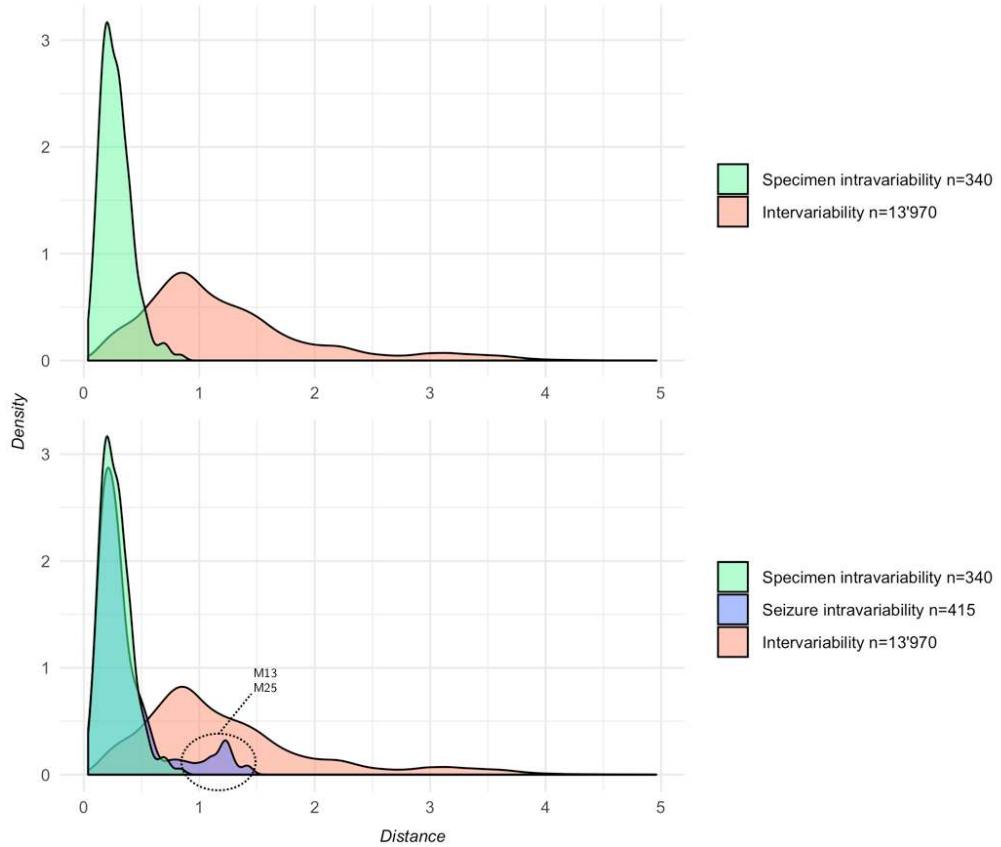


Figure 26: Density plots showing specimen-intravariability and intervariability (on top) and same representation including the seizure-intravariability (on bottom). Distance measurement values lying outside the specimen-intravariability are encircled with a dotted line.

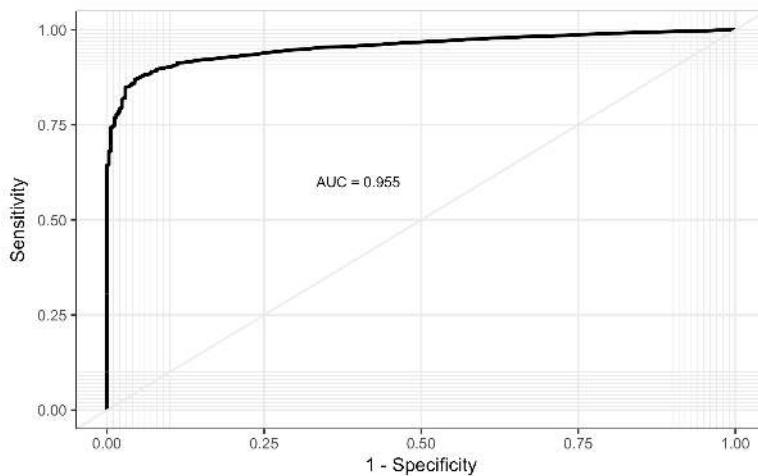


Figure 27: ROC curve and value for AUC for pXRF measurements

A threshold value had to be determined in order to cut the dendrogram resulting from the HCA. As already mentioned, the false negative rate should be as small as possible. A first possibility was again to choose the highest value of the specimen-intravariability

(distance = 0.855) as a threshold value. In this case, the sensitivity (TPR) would be at 1, whereas the specificity (TNR) would be 0.644. This means that, with such a model, no true positive link would be missed, but we would also have a false positive rate that would be 0.356, which is unreasonably high.

Another possibility is calculating the ‘optimal’ threshold, for which several methods exists. The Youden index J [130] is the point [x,y] on the ROC curve for which the distance to the line of equality is maximised. This means maximizing the difference between the TPR (sensitivity) and the FPR (1-specificity) [131]. By computing the Youden index, the optimal threshold value is at a distance of 0.53 with specificity = 0.953 and sensitivity = 0.872. Hence, for a minimal loss of specificity, a much better sensitivity could be gained. Figure 28 clearly illustrates the difference between the two cut-off points.

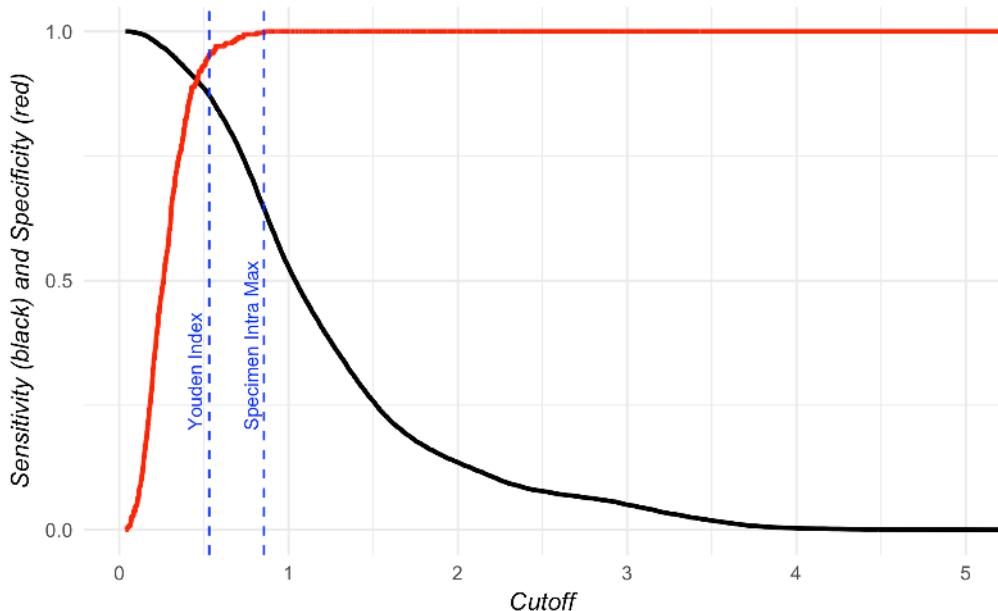


Figure 28: Sensitivity (true positive rate) in black and specificity (true negative rate) in red as a function of the cut-off distance. The dashed blue lines represent the threshold values that were set at the Youden Index and at the highest specimen-intravariability value.

5.2.4 Principal Component Analysis (PCA)

Figure 29 shows the PCA graph obtained for the pXRF results, the colours indicate the different watch models. Some specimens of the same model show a tendency for spatial closeness. For example, all specimens for each of the models Cosmograph

Daytona, Submariner (except for *M02*) and Datejust are close together. But no clear pattern or separation between specimens can be observed.

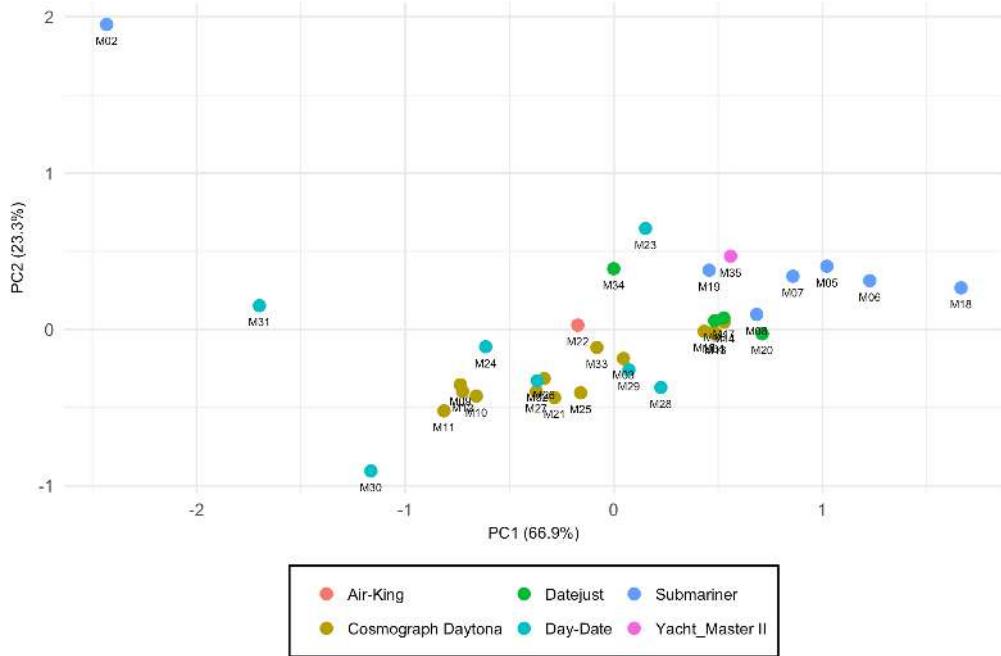


Figure 29: Repartition of the watchcases as a function of the first (PC1) and the second (PC2) principal component. The first two principal components express 90.2% of the total variance. The colors indicate the different watch models shown in the legend.

The contribution of each of the four selected variables, as well as a scree plot showing the variance attributed to each principal component can be found in Figure 30. The first two principal components explain 90.2% of the total variance, which is not surprising since only four variables were used.

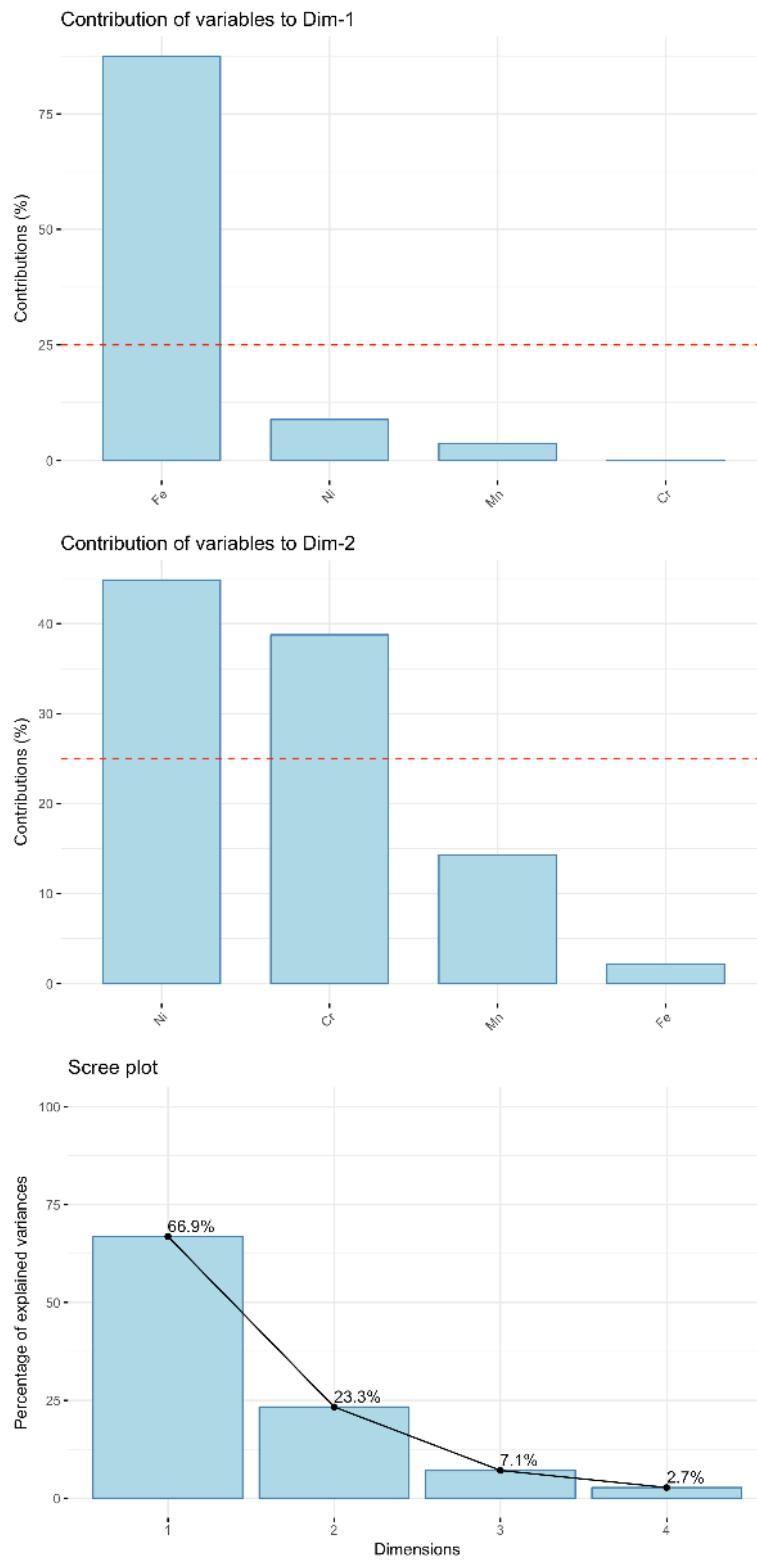


Figure 30: Bar plots of variable contribution to PC1 (on top) and PC2 (in the middle) and scree plot representing the percentage of explained variance of each principal component (on the bottom)

5.2.5 Hierarchical Cluster Analysis (HCA)

Each variable was defined by the mean value that was determined for the 5 replicate measurements. The applied similarity measure was the Euclidean distance and Ward's agglomeration method was used. The threshold defined using the Youden index yielded 14 groups and is represented in Figure 31 and Table 46; *M01* was added as group 15.

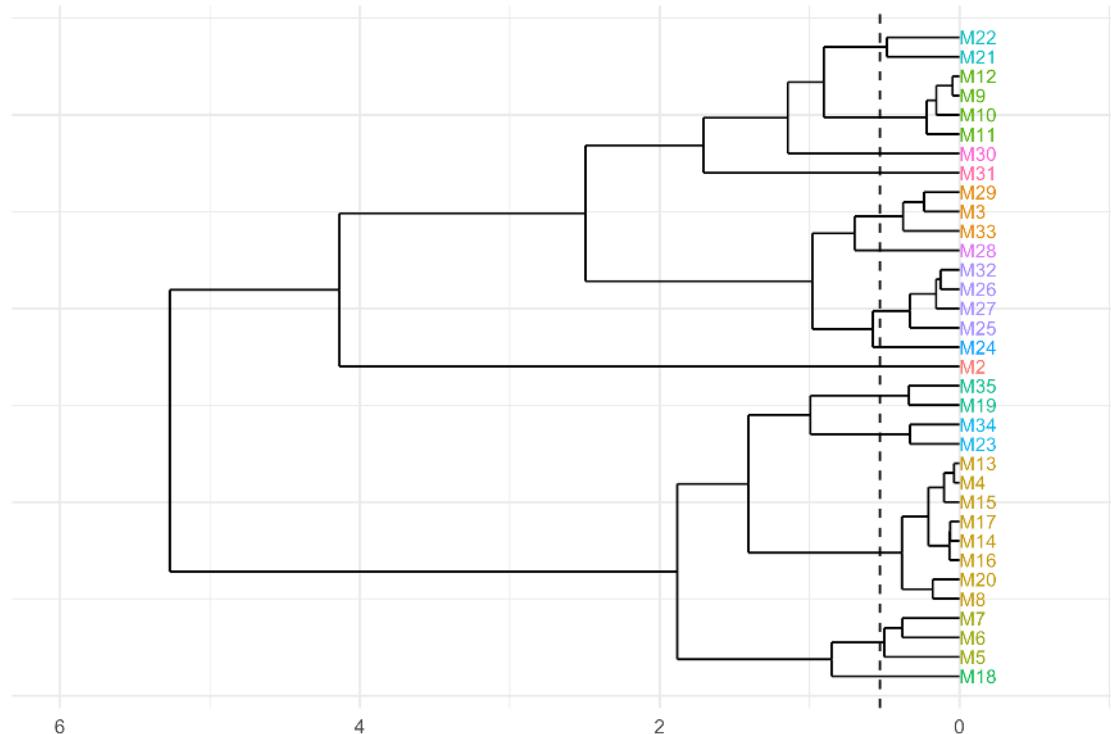


Figure 31: Dendrogram showing the results of the HCA, the black dashed line indicates the threshold that was used to form the 14 groups

Chemical profiling

| Group | Watch/ Model | Picture | Group | Watch/ Model | Picture |
|-------|----------------|---|-------|------------------|---|
| 1 | M22 Airking |  | 10 | M35 Yacht-Master |  |
| | M21 Daytona |  | | M19 Submariner |  |
| 2 | M12 Daytona |  | 11 | M34 Datejust |  |
| | M09 Daytona |  | | M23 Day-Date |  |
| 3 | M10 Daytona |  | 12 | M13 Daytona |  |
| | M11 Daytona |  | | M04 Daytona |  |
| 4 | M30 Day-Date |  | 13 | M15 Datejust |  |
| | M31 Day-Date |  | | M17 Datejust |  |
| 5 | M29 Day-Date |  | 14 | M14 Datejust |  |
| | M03 Daytona |  | | M16 Datejust |  |
| 6 | M33 Daytona |  | 15 | M20 Datejust |  |
| | M28 Day-Date |  | | M08 Submariner |  |
| 7 | M32 Day-Date |  | 13 | M07 Submariner |  |
| | M26 Daytona |  | | M06 Submariner |  |
| 8 | M27 Daytona |  | 14 | M5 Submariner |  |
| | M25 Daytona |  | | M18 Submariner |  |
| 9 | M24 Day-Date |  | 15 | M01 Deepsea |  |
| | M02 Submariner |  | | | |

Table 46: Watch specimens according to their chemical classes obtained through HCA using the pXRF results, including the watch model of each specimen and image of the watchcase

5.3 Comparison of grouping between ICP-MS and pXRF results

The analytical results from ICP-MS and pXRF both allowed for the specimens to be grouped based on their chemical resemblance. The intra- and intervariability distributions both indicated the possibility to distinguish between the populations. However, a comparison of the multivariate data analysis using the two results also showed that neither the spatial distribution of the specimens on the PCA plots nor the clustering on the dendrogram provided the same results. For example, the primary partition into the first three clusters is already quite different.

PCA using the pXRF data showed a slight tendency to group the watch cases according to their model. This can also be seen in Table 46, where several groups contain watches of the same model that are also visually similar. The PCA plot using the ICP-MS data shows that almost all the specimens are close together in a spatial sense, and the model grouping tendency is less pronounced.

As already outlined in section 4.7, the major elemental composition obtained by pXRF and ICP-MS do not agree; however, it cannot be said that the methods do not agree, since the measurements were not performed on the same location on the watchcases (surface vs. inner composition).

In chapter 7, the focus will be placed on the integration of the chemical information with the physical profiling of the counterfeit watches. For this purpose, the chemical profile determined by the ICP-MS results will be used. The objective of the contextualisation is to gain knowledge on the relationships that exist between the different steps of watch counterfeiting. It was initially assumed that the production origin can be reflected by the inner composition of the watchcases; hence, it was decided to consider the trace element profiles obtained by ICP-MS analysis for the contextualisation. The final chapter discusses whether pXRF, in our case, may or may not be a suitable alternative to ICP-MS as well as its application as a screening method for potentially toxic heavy metals.

6 Physical profiling

This chapter is devoted to the study of the links that can be drawn between the watches based on the codification of the physical characteristics of all imprints present on the considered watches.

The relationships that exist between the 35 watches of the specimen set, hereafter called ‘physical links’ were examined in the first place. A second source of information that was studied, were the connections from the 35 watches that were drawn with previously inspected watches, called ‘physical links on extended level’. Physical links on extended level were directly used from the lists that were generated in the expert reports.

All information on imprints present on the watches from the specimen set were retrieved with an SQL query from the relational database of the FH.

6.1 Study of the physical links

The total number of imprints found on the 35 watches was 489, each specimen containing between 9 and 18 imprints. In total, 243 different imprints were recorded. They are distributed as follows on the different components (Figure 32):

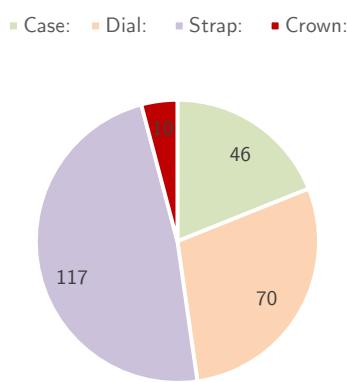


Figure 32: Distribution of individual imprints on watch components

Within these 489 imprints, 145 occurred only once, meaning that they did not generate a link with any other watch of the specimen set. This means that 98 codes were shared

at least by two of the 35 specimens, corresponding to a total of 344 occurrences within the 35 watches. Hence numerous physical links exist.

Nevertheless, these numbers do not reveal any information on how the 35 counterfeit watches are linked between each other on a physical level. They were handled through a network visualization as shown in Figure 33. Different colours were used to represent the different watch parts where the imprints are located. Only codes of imprints that generated links between watches are displayed, hence the 145 codes without any connections were omitted. If several codes on the same component were identified on the same watches, they were summarised, and the number of codes on the same component was added to the figure.

Physical profiling

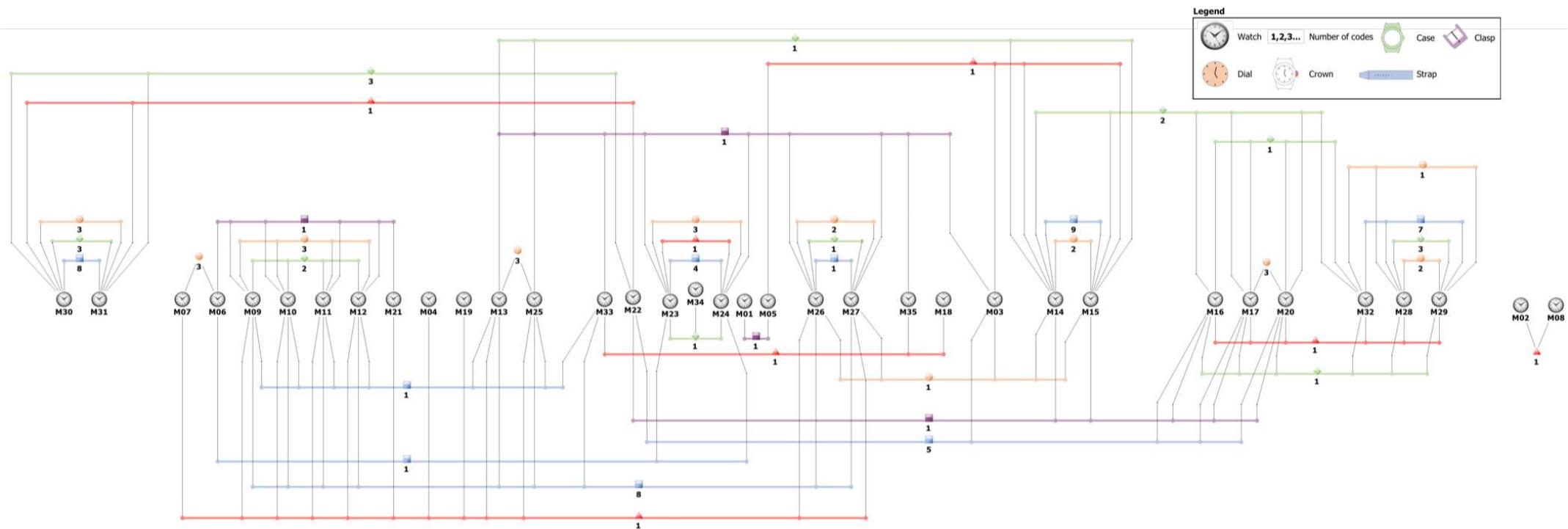


Figure 33: Representation of the physical links of imprints between the 35 counterfeit watches. Different colours were used to indicate the watch part where the imprints were found (green – case, purple – clasp, orange – dial, blue – strap and red – crown). The number of codes that form the same connections are represented by the black digits.

The visualisation does not provide a clear structure of linked imprints between the different watch parts. Nevertheless, among the 35 watches numerous links were identified. Specimens of a same model that were seized at the same time (e.g. M09/M10/M11/M12, M13/M25, M23/M24, M28/M29 and M30/M31) are connected between each other through all imprints. The remaining specimens share less connections between multiple components. Some watches are only linked through the imprints on the crown (represented in red) or the clasp (represented in purple) but do otherwise not share any physical connection between each other.

It was in a first step interesting to investigate the imprints that generated the most links. The top ten codes generated between 9 and 12 connections and are shown in Table 47. Most of the links were generated from imprints on the strap and on the crown. It was found that the code 502.0135.2019 (9 links) is an application of the brand logo on the clasp, which is part of the strap. Several categories of codes (brand, logo, quality related and source indication) can be found without however confirming a general trend.

| Code | Image | Part | Links |
|---------------|-------|---------------|-------|
| 304.0030.2260 | | Strap | 9 |
| 107.0001.0455 | | Strap | 9 |
| 304.0200.0062 | | Strap | 9 |
| 301.0094.0290 | | Strap | 9 |
| 301.0367.0289 | | Strap | 9 |
| 107.0010.0287 | | Strap | 9 |
| 106.0411.1414 | | Strap | 9 |
| 106.0211.0545 | | Strap | 9 |
| 502.0135.2019 | | Strap - Clasp | 9 |
| 102.0141.2092 | | Crown | 12 |

Table 47: Codes representing the top ten links between the 35 watches, representation of the imprints and watch part of the imprint

6.2 Study of physical links on extended level

In order to study the physical links on an extended level, the data had to be exempted from double values. This means, for example, if a specific code found on watch #1 was linked to 45 watches in the database, then watch #2 (analysed shortly after) had the same code, then the same 45 watches including watch #1 would appear. The question was however: How many links were generated by within the specimen set and within the entire database?

The total number of occurrences from the 243 individual imprints within the entire database and the number of links to all previously analysed watches was investigated. A number of 190 individual codes generated at least one link with another watch. All these codes together generated links between 504 watches that were analysed previous to the specimens of our *specimen set 1*. Between the 35 watches, 243 individual codes were identified, meaning that 53 codes had no anteriority recorded in the database.

Several of these 190 codes generated links with only a small number of formerly analysed watches in the database. We can reasonably expect that imprints generating a multitude of links are more likely to inform on the structure of the market. In addition, codes that generate several links indicate the use of the tagging tool for mass production. It should be noted that all codes representing an imprint found on the crowns of the 35 initial specimen set were linked to formerly analysed imprints located on crowns, whereas imprints on other components were not always recorded before.

We decided to keep our sights on codes that had already been found several times. More precisely, codes that had links with at least 25 others were selected, extracted and further investigated. The information about their location on the watch components can be found in Table 48.

| | # codes | # occurrences in DB | # of watches |
|-------------------------------------|-----------|---------------------|--------------|
| Total number of codes with links: | 190 | 2124 | 504 |
| # of codes with more than 25 links: | 27 | 1129 | 365 |
| - on strap: | 23 | 973 | 312 |
| - on crown: | 3 | 111 | 111 |
| - on case: | 1 | 45 | 45 |
| - on dial: | - | - | - |

Table 48: Total number of codes connected to at least one other watch in the database, number of codes connected to 25 or more watches and their distribution according to the different watch components

From the initial 190 codes, it was found that retaining only the 27 codes with more than 25 links represent more than half of the total number of occurrences (2124 vs. 1129). Furthermore, imprints located on straps and crowns account for 96% of the total number of occurrences.

A further differentiation could be envisaged between the content of information represented by the imprints. They can be distinguished as follows:

- *brand-specific*, e.g. the name or the logo of the brand
- *model-specific*, e.g. the name of the model or model identification number
- *non-specific*, e.g. technical and geographical indications

Their classification seems important, since they convey different information. Model-specific imprints can only generate links between watches of the same brand and the same model. Two watches of a common brand but different models could not share

model-specific imprints, even though they possibly originate from a common production line. Brand-specific imprints are logically found on watches of the same brand. Non-specific imprints are very interesting, since they could possibly be used on all counterfeit watches, even on those of different brands. Brand-specific and non-specific imprints are thus more appropriate to gather information on potential mass production. In this respect, it became apparent that the vast majority of the 27 considered imprints were either brand- or non-specific, whereas model-specific imprints were only found three times. Table 49 provides an overview on these imprints, including information on their specificity, location as well as the number of links found in the database. For reasons of simplification, all imprints were labelled with short ID numbers.

Physical profiling

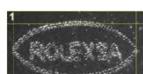
| ID | Code | Image | Specificity/ Part | Links | ID | Code | Image | Specificity/ Part | Links |
|-----|---------------|---|-------------------------|-------|-----|---------------|---|-------------------------|-------|
| #1 | 107.0010.0287 |  | None Strap | 25 | #15 | 301.0367.0257 |  | Brand Strap | 40 |
| #2 | 107.0001.0455 |  | None Strap | 25 | #16 | 305.1464.0035 |  | None Strap | 41 |
| #3 | 107.0010.0154 |  | None Strap | 25 | #17 | 102.0141.2092 |  | Brand Crown | 43 |
| #4 | 106.0411.1414 |  | None Strap | 25 | #18 | 105.2437.0009 |  | None Strap | 44 |
| #5 | 106.0211.0545 |  | None Strap | 25 | #19 | 105.2408.0007 |  | Model Strap | 44 |
| #6 | 301.0367.0289 |  | Brand Strap | 25 | #20 | 301.7624.0030 |  | Model Case | 45 |
| #7 | 301.0094.0297 |  | Brand Strap | 28 | #21 | 101.0094.1854 |  | None Strap | 47 |
| #8 | 102.0141.2100 |  | Brand Crown | 32 | #22 | 305.7181.0003 |  | None Strap | 56 |
| #9 | 304.0229.0048 |  | None Strap | 32 | #23 | 106.0211.0449 |  | None Strap | 64 |
| #10 | 301.0094.0180 |  | Brand Strap | 32 | #24 | 107.0001.0421 |  | None Strap | 64 |
| #11 | 301.0094.0283 |  | Brand Strap | 33 | #25 | 106.0411.1352 |  | None Strap | 66 |
| #12 | 102.0141.2018 |  | Brand Crown | 35 | #26 | 301.0367.0110 |  | Brand Strap | 69 |
| #13 | 502.0135.1771 |  | Brand Strap Clasp | 35 | #27 | 502.0135.2019 |  | Brand Strap Clasp | 90 |
| #14 | 305.3367.0004 |  | Model Strap | 38 | | | | | |

Table 49: Imprints (including ID and code number) that generated more than 25 links, their specificity, watch part and number of links observed. Colour codes are assigned to groups of imprints that were mostly found in combination on the same watches.

The following groups of imprints were generally found in combinations on the same watches:

- Codes # 1,2,4,5 and 6 were found on 25 watches, called imprint group 1
- Codes # 9,11,14,15 and 16 were found on 31 watches, called imprint group 2
- Codes # 18,19,21 were found on 43 watches, called imprint group 3
- Codes # 22,23,24,25 and 26 were found on 52 watches, called imprint group 4

These four groups are composed of imprints that were found on the straps. Codes belonging to different groups were never found on the same watch, hence there is no mixing between these four groups.

The code that generated the most links is #27. This code corresponds to an imprint with the brand logo in form of an application that is attached to the clasp. This code is found on 90 watches, but within several groups of codes and not found in specific combinations. Non-specific imprints are mostly found to be quality indications rather than geographical or technical indications.

A visualisation of the links presented in Table 49 can be found in Figure 34. Several simplifications were made to present a more legible picture. The three codes corresponding to the crowns (#8, #12, #17), as well as the two codes corresponding to the clasp application were (#13, #27) were discarded. Within the identified imprint groups, not all watches presented the total number of imprints included in these groups but always a combination of several imprints. Nevertheless, these specific combinations were omitted. This again allowed a clearer representation and the four imprint groups can be clearly identified. Watches were separated according to their country of seizure which allowed to represent the transnational spread of the linked specimens. In total, 267 watches are represented by these 22 imprints, from which 21 are found on the strap and 1 on the case.

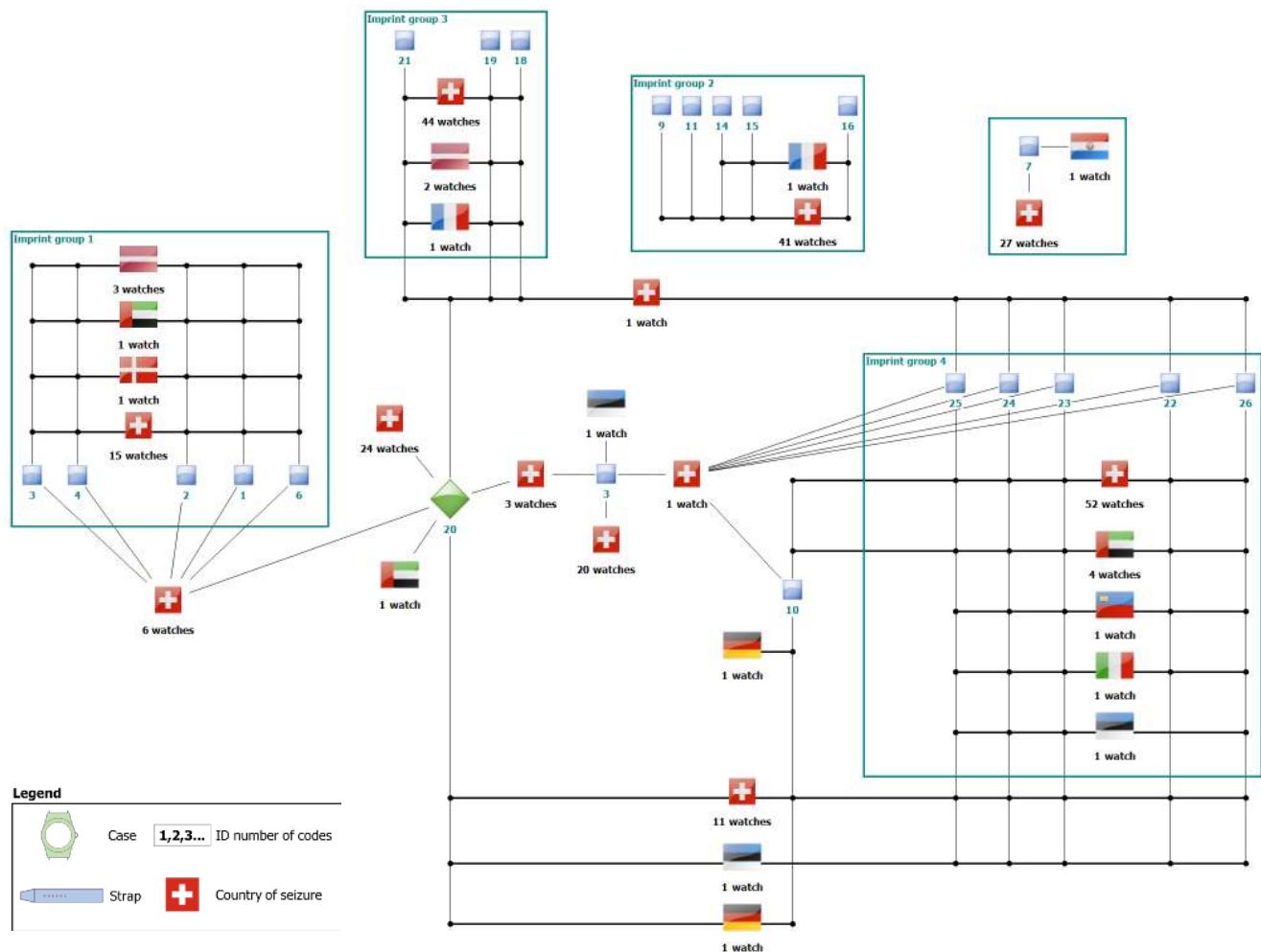


Figure 34: Representation of the links obtained by the selected 22 imprints (strap and case) with watches analysed previous to the initial specimen set. The identified groups of imprints that were generally found in combination are highlighted by turquoise rectangles and the linked watches are separated according to their country of seizure.

Physical profiling

Among all imprints there were 145 codes that did not generate any connection within the 35 watches. However, after analysing all codes within the entire database, only 52 had no recorded anteriority in the database. It is important to consider these imprints, since the tools that generated them are less likely to reveal production on an industrial scale. These 52 codes are composed of 16 imprints on the case, 15 on the dial and 21 on the strap and can be found in Table 50.

| Code | Image | Specificity/ Part | Code | Image | Specificity/ Part |
|---------------|-------|----------------------|---------------|-------|----------------------|
| 301.3103.0028 | | Model <i>Case</i> | 305.3872.0001 | | Model <i>Case</i> |
| 301.3107.0007 | | Model <i>Case</i> | 301.0629.0001 | | Model <i>Case</i> |
| 301.3292.0002 | | Model <i>Case</i> | 401.0313.2253 | | Model <i>Dial</i> |
| 605.1447.0004 | | Model <i>Case</i> | 401.0316.2279 | | Model <i>Dial</i> |
| 301.3609.0001 | | Model <i>Case</i> | 401.0731.0292 | | Model <i>Dial</i> |
| 305.4148.0005 | | Model <i>Case</i> | 401.3124.0020 | | Model <i>Dial</i> |
| 301.8112.0012 | | Model <i>Case</i> | 404.0030.5096 | | None <i>Dial</i> |
| 302.0094.0066 | | Brand <i>Case</i> | 401.0341.2295 | | Model <i>Dial</i> |
| 305.1559.0001 | | Model <i>Case</i> | 404.0030.5142 | | None <i>Dial</i> |
| 107.0009.0101 | | None <i>Case</i> | 404.0030.5193 | | None <i>Dial</i> |
| 305.2515.0038 | | None <i>Case</i> | 404.0030.5204 | | None <i>Dial</i> |
| 301.0596.0001 | | Model <i>Case</i> | 401.3099.0014 | | Model <i>Dial</i> |
| 305.3134.0020 | | None <i>Case</i> | 405.0905.0017 | | None <i>Dial</i> |
| 305.3208.0004 | | Model <i>Case</i> | 401.0313.2254 | | Model <i>Dial</i> |

Physical profiling

| Code | Image | Specificity/ Part | Code | Image | Specificity/ Part |
|---------------|-------|-----------------------|---------------|-------|-----------------------|
| 404.0030.5199 | | None <i>Dial</i> | 305.0320.0004 | | None <i>Strap</i> |
| 404.0030.5188 | | None <i>Dial</i> | 305.1464.0085 | | None <i>Strap</i> |
| 405.0315.1292 | | None <i>Dial</i> | 305.3243.0016 | | None <i>Strap</i> |
| 502.0135.2057 | | Brand <i>Strap</i> | 104.1117.0020 | | Model <i>Strap</i> |
| 101.0094.1923 | | Brand <i>Strap</i> | 502.0135.2055 | | Brand <i>Strap</i> |
| 102.0135.2162 | | Brand <i>Strap</i> | 301.0094.0439 | | Brand <i>Strap</i> |
| 301.0367.0434 | | Brand <i>Strap</i> | 301.0367.0437 | | Brand <i>Strap</i> |
| 301.0367.0439 | | Brand <i>Strap</i> | 302.0188.0015 | | Model <i>Strap</i> |
| 105.3120.0004 | | Model <i>Strap</i> | 304.0229.0101 | | None <i>Strap</i> |
| 302.0167.0004 | | Model <i>Strap</i> | 305.1464.0086 | | None <i>Strap</i> |
| 302.0200.0002 | | Model <i>Strap</i> | 305.3053.0047 | | Model <i>Strap</i> |
| 107.0010.0346 | | None <i>Strap</i> | 502.0135.2059 | | None <i>Strap</i> |

| | |
|---------|---------------------------|
| 4 codes | Links between M23/24 |
| 2 codes | Links between M26/M27 |
| 7 codes | Links between M28/M29/M32 |

Table 50: Codes that did not generate any links in the database, the specificity regarding brand or model and the watch part where the codes were found. Different colours were used to indicate imprints that generated links between watches of the specimen set, as shown in the legend.

Within these codes, 9 are brand-specific, 25 are model-specific and 18 non-specific. In total, 39 codes had no anteriority recorded in the database and were found for the first time on a watch from the specimen set. However, 13 imprints were found to link some watch pairs from the specimen set.

6.2.1 Temporal analysis of imprints

The expert reports issued for the 35 watches contained not only the connections to previously analysed watches but also some temporal information. The date that was used for the temporal analysis corresponded to the beginning of the physical examination at the FH, which is relatively close to the date of seizure. In this way it was possible to investigate the evolution of the imprints that generated the most connections. Even if the production time remains unknown, the appearance and persistence of imprints can be studied. Figure 35 illustrates the four groups of imprints found in combination, as explained above (see section 6.2). Consequently, these codes also exhibit the same temporal behaviour. Codes with no specific relation to others are displayed in Figure 36.

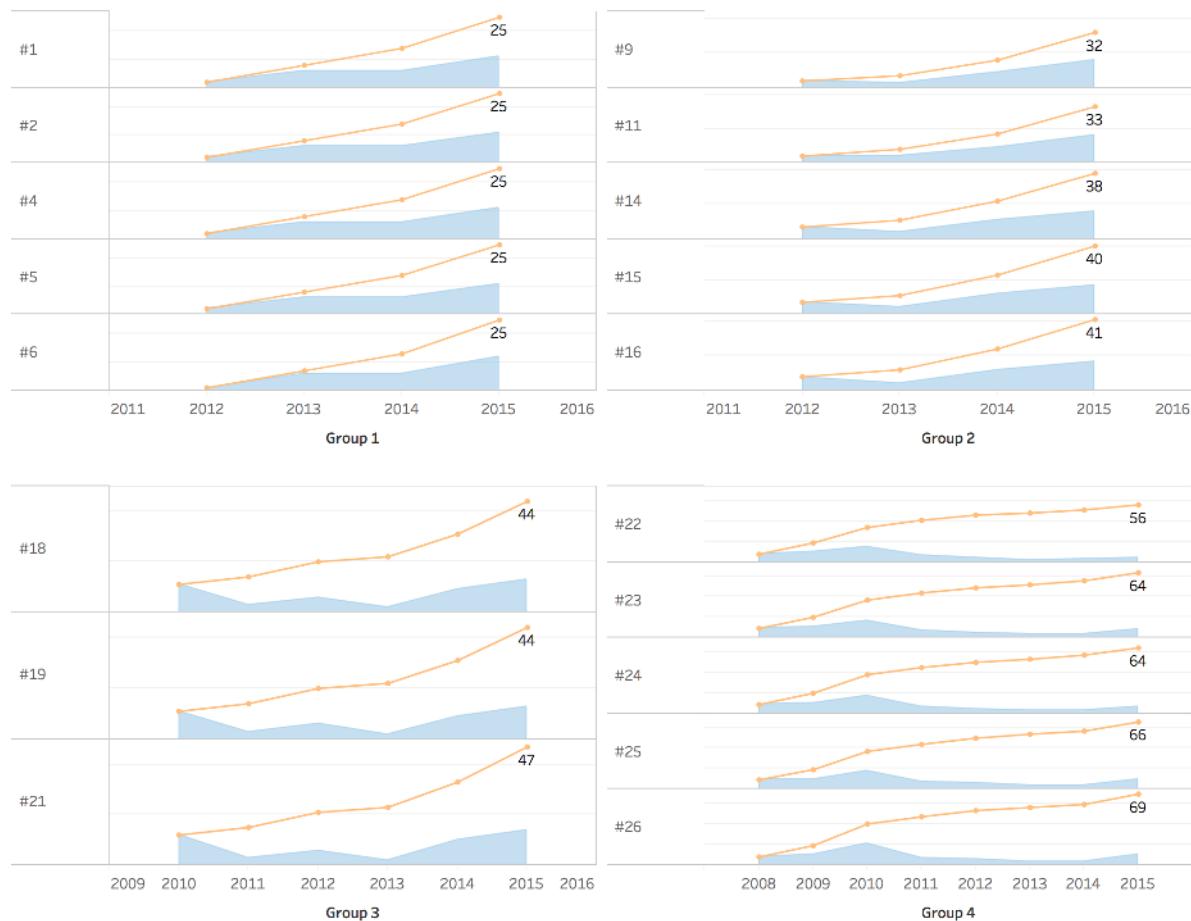


Figure 35: Temporal analysis showing the number of links/code/year. Codes are grouped according to those found in combination on the same watches.

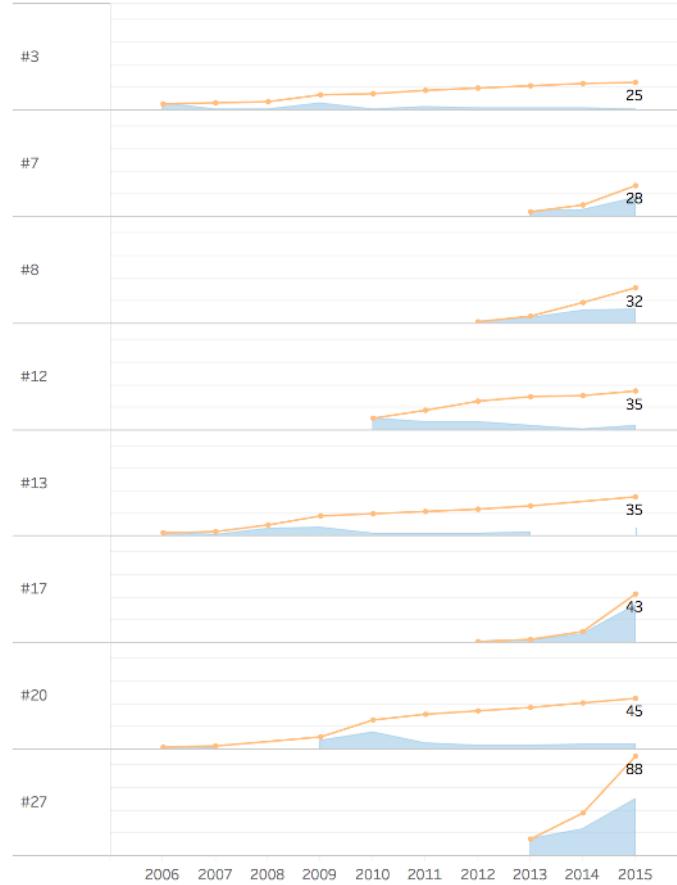


Figure 36: Temporal analysis showing the number of links/code/year. The codes displayed in this figure were not found in specific combinations with others.

The codes #3, #13 and #20 persisted over a period of almost 10 years. Code #3 generated links with 90 other watches but was found for the first time only 2 years prior to the analysis of the watches contained in the specimen set.

6.2.2 Geographical spread

The information retrieved for the geographical spread was performed on all 504 watches, which means all the watches that were linked via at least one characteristic to the 35 watches of the original specimen set. Even though the vast majority of the watches were seized at Swiss borders, some watches were being seized during actions taken in 13 other countries on three continents (visualised in Figure 37). It has to be kept in mind, that the country of seizure is not the country of production. Very little is known on the provenance and transit of these watches. Yet, the countries of seizure demonstrate the spread of the watches around the globe once they had been sold.

Physical profiling

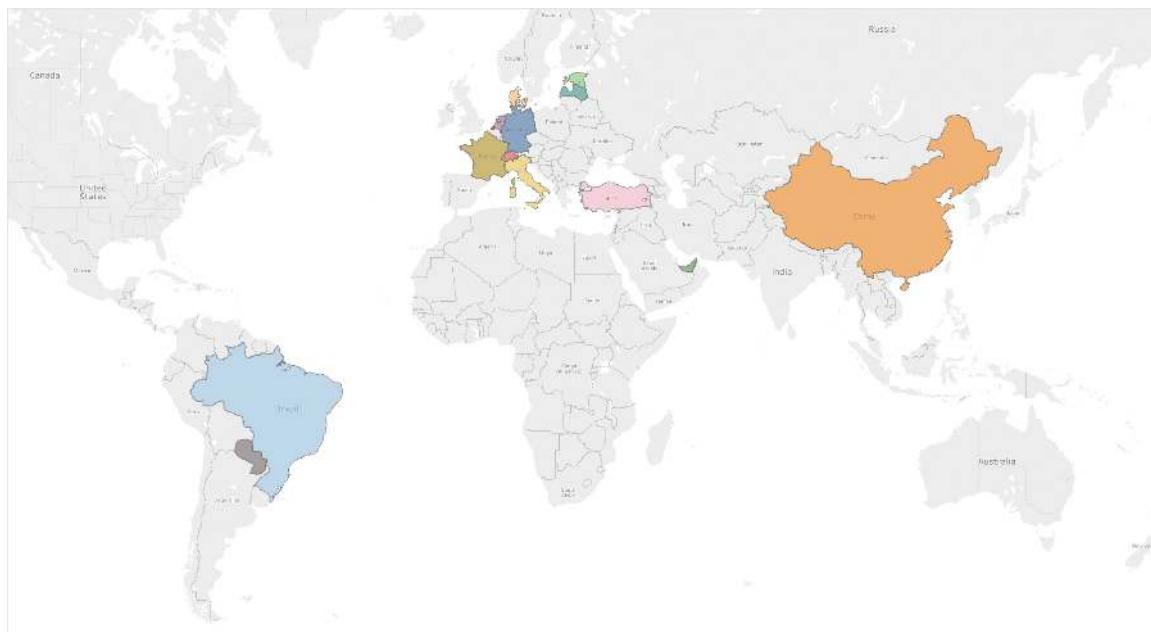


Figure 37: Countries of seizure for the 504 watches linked through imprints contained on the watches from the initial specimen set

7 Combination of chemical and physical links

The goal of this chapter was to confront the chemical and physical links between the 35 counterfeit watches and to investigate the relationships that exist among them.

As seen in Chapter 5, the chemical profiles that were retained consisted of 10 variables (scenario 1) from the minor elements measured by ICP-MS. On the basis of these profiles, 15 groups were formed by HCA. We assumed that the ICP-MS results better reflect the watchcase production than the results obtained by pXRF, which is a surface analysis technique. The developed analytical strategy based on ICP-MS allowed to access the inner steel composition without considering the surface layer. The completion of a watchcase may include additional plating steps, hence using XRF results could lead to misinterpretation if we search for links that reflect the watchcase production origin.

As seen in Chapter 6, the physical links between imprints need to be considered on each component separately. The codes that were found in combination were merged into groups to simplify the visualisation. As previously stated, analysis of the physical links allowed to observe that the imprints on the crowns and on the clasps have generated links between watches that otherwise were not linked on a physical level. The codes representing those imprints were also discarded to illustrate a more legible picture of the different connections. Figure 38 shows a visual representation of the chemical and physical links between the 35 counterfeit watches. Different colours were used to represent the different watch parts where the imprints are located. The watch specimens are displayed in the middle, represented by a watch icon, chemical connections can be found in the upper part of the figure, whereas the physical relationships are shown on the lower side.

Combination of chemical and physical links

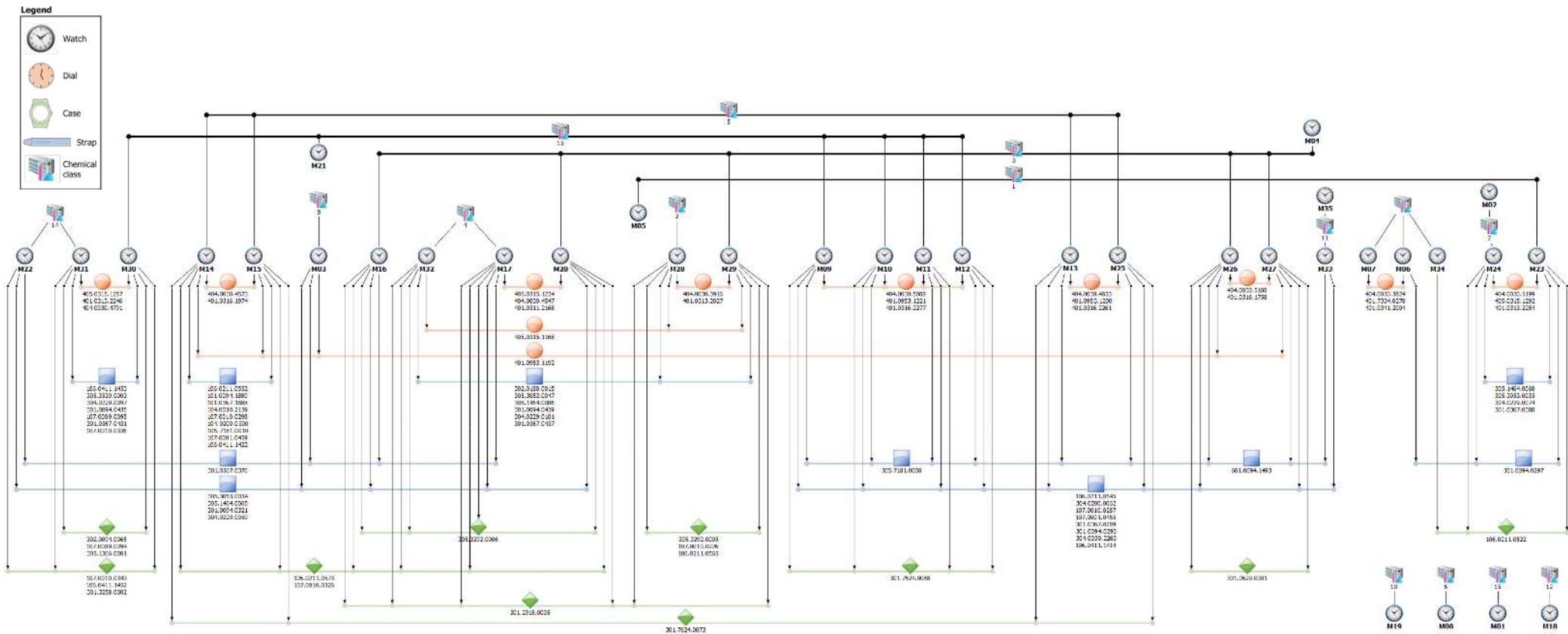


Figure 38: Visualisation of the combination of chemical and physical links between the 35 counterfeit watches. Different colours were used to indicate the watch part where the imprints were found (orange – dial, green – case and blue – strap). The codes that form the same connections are listed below each other.

Overall, numerous connections between almost all specimens can be found on a chemical and physical level. Generally, there is not much correspondence between the chemical and physical links. However, some chemical links were confirmed by physical links between the entire set of imprints present on all components. In such cases, the watches were visually indistinguishable and were seized simultaneously (e.g. M09/M10/M11/M12, M14/M15, M13/M25 and M26/M27). Specimens M01, M08, M18 and M19 do not share chemical or physical links on the straps dial or case with any other watch (clasps and crown excluded).

On the one hand it was possible to observe watches that belong to the same chemical group but without links between their imprints. This holds for specimens M06/M34, M02/M24 and M33/M35, but the pair is linked through two imprints on the crown and the clasp respectively, which were discarded in this representation. On the other hand, physical links were not necessarily confirmed by chemical ones, e.g. M23/M24 and M30/M31. Another interesting example was found in the relationships between specimens M28/M29/M32 shown in Figure 39.

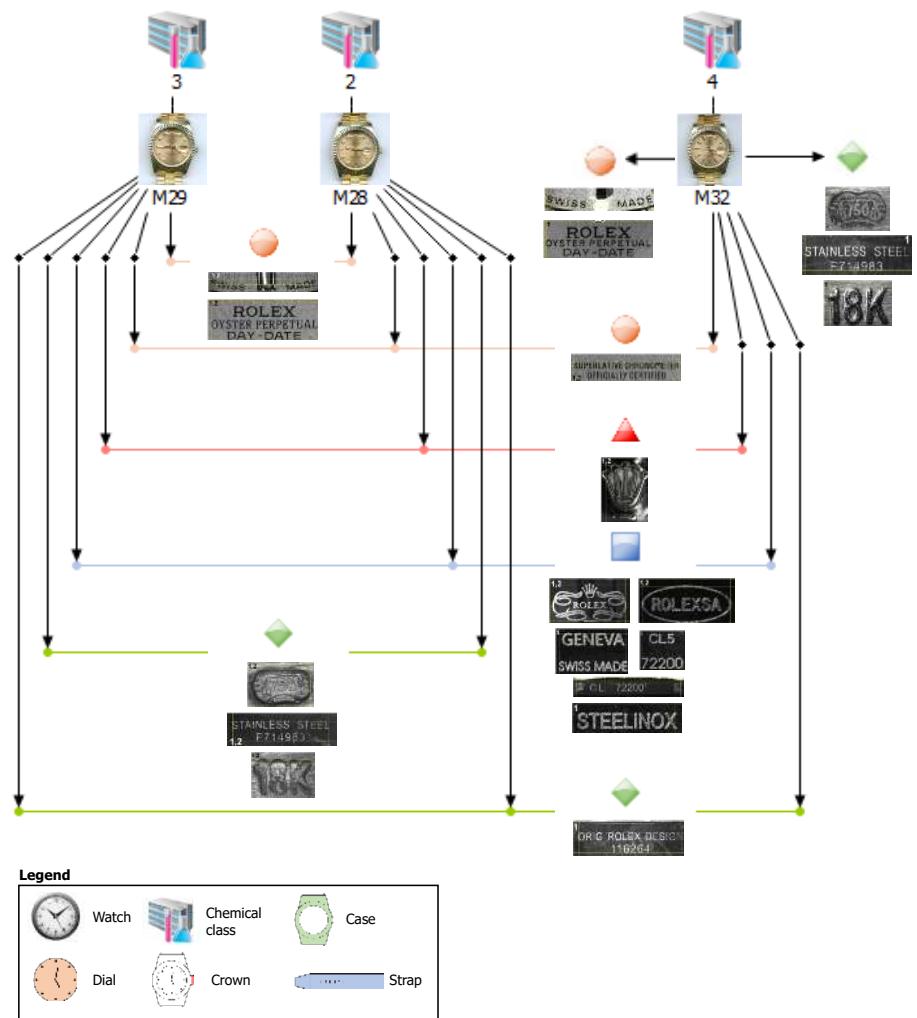


Figure 39: Visualisation of chemical groups and the physical links between the imprints of specimens M28/M29 and M32

These three specimens are gold plated counterfeit Rolex watches of the same model (Day-Date). Visually, no major differences can be perceived between them. Yet, the watches were not found to be chemically related. Specimen pair M28/M29 was initially considered to be part of the intra-variability. Distance measurements between the selected variables revealed values far above the chosen threshold. This also applies to the distance measurements between M28/M32 and M29/M32, even if the comparison between the latter specimen pair showed values close to the threshold. Considering the physical links, it was found that 9 out of 14 imprints are linked between the 3 specimens. The 5 imprints that are not linked (2 on the dial and 3 on the strap) are, however, of the exact same type. It was thus interesting to further compare these imprints, since some of them consist of several parts or lines of text and could possibly

be tagged independently. An attempt was made to superimpose these non-corresponding imprints (Table 51).

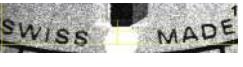
| Imprint M28/M29 | Imprint M32 | Comparison | Comments |
|---|---|---|--|
|  |  |  | Possible to superimpose 'SWISS' and 'MADE' independently |
| 404.0030.3916 | 404.0030.5193 | | |
|  |  |  | Possible to superimpose 'ROLEX' Different fonts for 'OYSTER PERPETUAL' Different size for 'DAY-DATE' |
| 401.0313.2027 | 401.0313.2253 | | |
|  |  | - | Visually distinguishable |
| 107.0010.0226 | 107.0010.0326 | | |
|  |  |  | Possible to superimpose 'STAINLESS STEEL' and 'F714983' independently |
| 305.3292.0005 | 305.3292.0006 | | |
|  |  | - | Visually distinguishable |
| 106.0211.0565 | 106.0211.0573 | | |

Table 51: Superimpositions of non-linked imprints between M28/M29 and M32. Imprints consisting of multiple parts were overlaid separately if necessary.

First of all, it was possible to overlay the 'SWISS MADE' imprint when considering the two parts independently. The second pair 'ROLEX OYSTER PERPETUAL DAY-DATE' it was possible to match the 'ROLEX' part. The second line ('OYSTER PERPETUAL') consists, however, of a different font type, and the last part ('DAY-DATE') is of a different size. The fourth pair with the inscription 'STAINLESS STEEL F7129' nearly matched. It is possible that the surface to which these imprints were tagged on are not flat and that distortions are possible (especially on parts like the case where the surface is often not flat). This may lead to incorrect differentiations between imprints. The third and fifth pair were visually distinguishable. It is important to mention that the superimpositions were made without any measurements of the imprints. The images of the imprints were used directly from the expert reports and

the size was manually adapted to fit between the two images. Nevertheless, this approach can be justified since only potential matches were sought and the resulting information will solely be used to discuss potential consequences.

As a final point, it was worth examining the chemical and physical relationships on the watchcases since it was assumed, that the results from ICP-MS analysis reflect the watchcase production. These connections are displayed in Figure 40. It can be seen, that several links exist among the watchcases on a physical and chemical level. Nevertheless, several specimens that belong to independent chemical groups and that do not share any links between imprints on the cases (e.g. M19, M03, M18, M08, M01) were found. Several chemical links were also confirmed by physical links or vice versa (e.g. M09/M10/M11/M12, M22/M31, M13/M25/M24/M25, M17/M32, M16/M20, M26/M27). There are also links between imprints on the cases belonging to watches that are chemically not related, e.g. M34/M24/M23.

Most links are generated from the code 301.2918.0005, linking 6 watches belonging to 3 chemical groups. The combination of the codes 107.0010.0326 and 106.0211.0573 was also found on 6 watches belonging to 3 chemical groups. These specific codes and the watch specimens on which they were found were examined in closer detail (see Table 52).

Combination of chemical and physical links

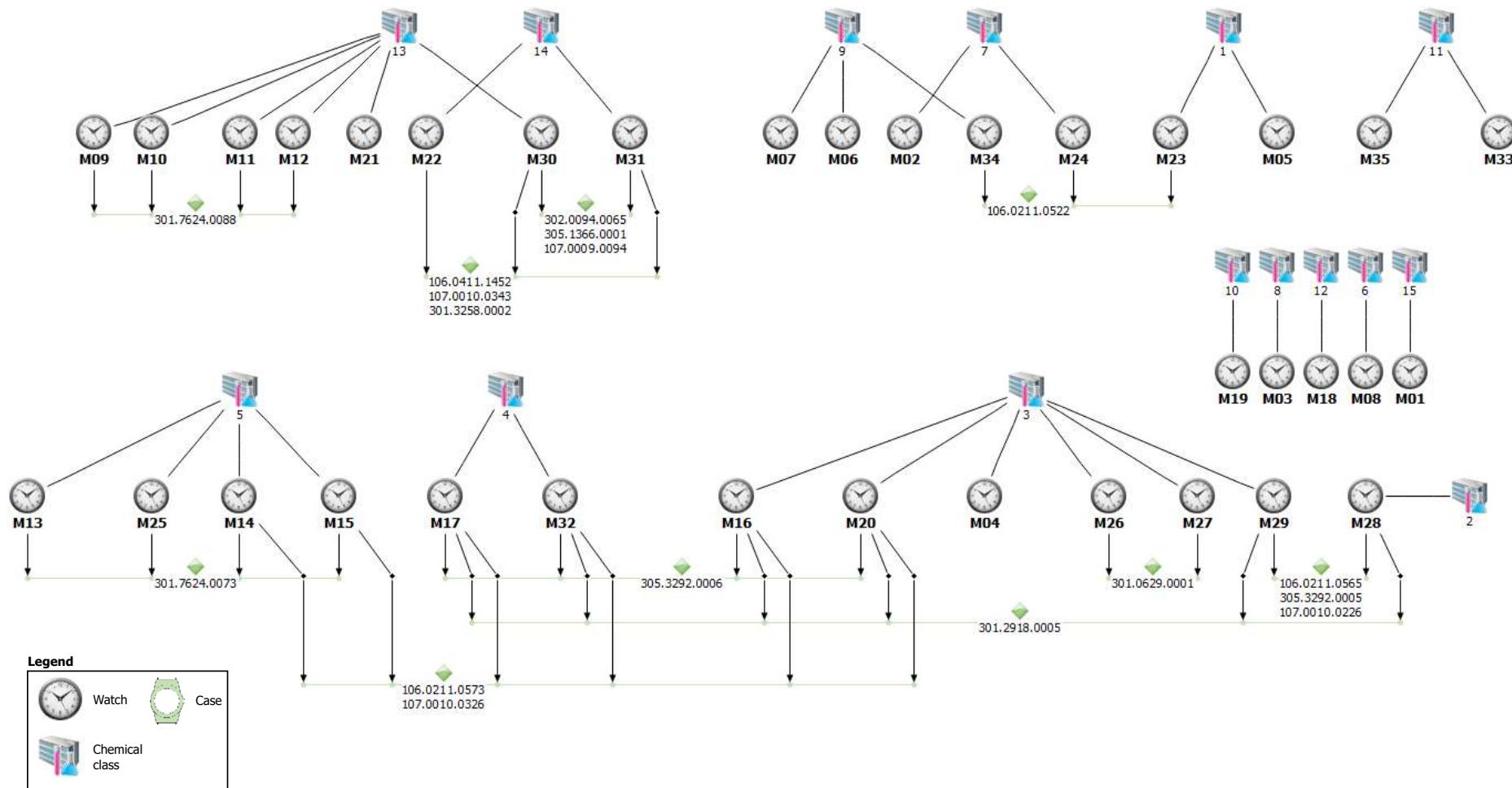


Figure 40: Visualization of chemical and physical links (considering only the imprints on the cases) between the 35 watches

Combination of chemical and physical links

| Code | Imprint | Watches | | Chemical groups |
|---------------|---|--|--|--|
| 301.2918.0005 |  | M17  2 | M32  1 | Group 4 |
| | | DATEJUST | DAY-DATE | |
| | | M16  1 | M20  2 | M29  2 |
| | | DATEJUST | DATEJUST | DAY-DATE |
| 107.0010.0326 |  | M28  1 | | Group 2 |
| | | DAY-DATE | | |
| | | M14  1 | M15  2 | Group 5 |
| | | COSMOGRAPH DAYTONA | COSMOGRAPH DAYTONA | |
| 106.0211.0573 |  | M17  2 | M32  1 | Group 4 |
| | | DATEJUST | DAY-DATE | |
| | | M16  1 | M20  2 | Group 3 |
| | | DATEJUST | DATEJUST | |

Table 52: Example of imprints present on multiple watches that belong to different chemical groups

It can be seen that imprints that were tagged with the same tool can be found on counterfeit watches of different models and different surface plating.

8 Discussion

Counterfeit watches are the product of an illicit activity. Investigative methods often focus in a reactive way to single seizures, which results in a limited vision of what is usually a far broader criminal network. It is necessary to emphasize the role of counterfeit goods as physical objects in order to understand specific aspects of the counterfeiting process; notably, the production and distribution on of the counterfeit goods and hence parts of the organizational structure that produced the counterfeits. The main insights acquired through this research are based on the exploitation of material traces. The information provided by these different types of traces allow us to distinguish or regroup specimens by measuring the proximity of their chemical compositions, the presence or absence of common physical features as well as circumstantial information. Over the previous chapters, a methodology for the chemical profiling of counterfeit watchcases was proposed. Additionally, new ways to exploit the information obtained from the physical profiling of imprints present on these watches were detailed. In the final stage, the two sources of information were combined. This chapter is devoted to the discussion of each of the initially proposed hypotheses and the methodological decisions that have been taken over the course of this thesis. Furthermore, a general discussion around the significance of the obtained results and the utility of forensic intelligence towards a deeper understanding of this particular illicit market is proposed. Finally, this chapter closes with the limitations and perspectives of this research.

8.1 Discussion on the selected specimen sets

It should be noted that the presented results suffer from certain inherent biases since the specimens obtained for investigation were entirely dependent on a few things: first and foremost, they were dependent on the seizures that were made by customs²⁵; secondly, on the specimens that were then retained by the FH for further physical examination; and finally, the specimens that were ultimately chosen for this research. To expand on the above-mentioned biases, the forensic intelligence process fundamentally relies on the detected material traces. Hence, the results will, to some extent, reflect ongoing strategies adopted by the authorities as a response towards counterfeiting. For example, if customs specifically target tourist traffic coming from Thailand or small postal parcels originating from China over a certain period of time. In such cases, it would be more likely to access counterfeit goods that originate from the same manufacturing network, which would not be representative of a phenomenon that is far more global.

Thus, the question arises if the physical evidence that was obtained and analysed over the course of this research reflects the entire illicit market for counterfeit watches. This can seriously be questioned considering that 35 specimens were analysed in this work, while globally over 1,920,000 counterfeit watches were seized in 2017 [47].

One of the limiting factors of this work was the limited specimen size. It was intentionally kept small because it would have been quite difficult to test and develop an analytical strategy with a large specimen set. Furthermore, an additional goal was to demonstrate that a vast amount of information could be gained from the chemical profiling of watchcases in combination with physical profiling associated to former links using just a small set of specimens. This issue will be further discussed in section 8.3 and 8.5.

The consideration of only a single brand for study could also be raised as a point of concern. Nevertheless, this choice is justified in our opinion, since it would be more

²⁵ Specimen set 1 was the primary one used for this thesis and consisted of watches that were seized by customs authorities in Switzerland. Nevertheless, counterfeit watches could also be obtained from other sources (e.g. seizures of small postal packages or seizures from large scale police actions in assembly sites or warehouses). Another alternative, independent of any interventions by the authorities, would be to physically purchase counterfeit watches or to order them online.

probable to gather meaningful information on the chemical and physical relationships between watches of a common brand.

In light of these limitations, it is important to interpret the obtained results critically and be aware that it is not possible to draw generalized market conclusions. Nonetheless, the proposed approach based on the triangulation of information related to traces of the production and distribution of counterfeit watches holds the potential to provide valuable knowledge on this illicit market.

8.2 Analytical approach and method comparison

Watchcases were selected as the objects of interest for the chemical profiling part. Numerous potential analytical approaches could be applied to measure the elemental composition of the alloys used to form the cases. The chosen approach, using XRF and ICP-MS, was inspired by previous applications in the field of jewellery analysis, where their use has already been successfully applied and documented. Going forward, the advantages of each method as well as the drawbacks and the challenges encountered will be detailed.

The most demanding part of ICP-MS analysis is the sample preparation. For our measurements, electrical discharge mining was used to obtain sample cuts from the watchcases. To prepare the samples for cutting, the watches first needed to be disassembled. This step usually required specialized watchmaking tools to open the back casing and retrieve the mechanical components. The sample cuts were then completely dissolved in order to be injected into the ICP-MS system. This step involved the use of strong acids, namely concentrated HCl and HNO₃. Given the highly corrosive nature of these chemicals, the use of proper methods for their storage, use and disposal is very important. Furthermore, these acids can react strongly with certain metals and release highly flammable hydrogen gas; care needs to be taken to use the proposed microwave-assisted acid digestion method exclusively for steel alloys²⁶. Nevertheless, watches of poorer quality may be made of alloys other than steel; in these cases, the dissolution matrix has to be adapted.

²⁶ It is absolutely necessary to perform a screening step to identify the type of alloy.

The major drawbacks of the ICP-MS method are the destructiveness of the method. In addition, although ICP-MS analysis is rather fast on its own, the sample preparation procedure is extremely time-consuming; and it requires a two-stage dilution to analyse the major and trace elemental groups.

Despite these drawback, the selected validation criteria fulfilled the analytical requirements with regards to the selectivity, accuracy, precision and had detection limits that were lower than the levels of the measured trace elements. Hence, we can **affirm the first hypothesis of this research, which stated that the analytical approach** (in terms of ICP-MS analysis) **allows us to obtain reliable data on the chemical composition of watchcases.** To the best of our knowledge, the application of ICP-MS for the analysis of major and trace elements in counterfeit watchcases is unique.

The analytical results offered a range of information on the watchcases from our specimen set. First, it was possible to formally identify the type of alloy. Second, we were able to accurately determine the trace elements present in the steel composition. Knowledge of the elemental composition allowed us to identify the raw material used for the production of the counterfeit watchcases; whereas, until now, information on counterfeit watches was primarily physical in nature. Furthermore, this type of data is independent of the brand that has been counterfeit.

For the development of the analytical method, the number of specimens ($n=35$) as well as the number of sample cuts ($n=175$) analysed were appropriate. Sufficient data could be gathered to demonstrate that the proposed methodology is suitable to measure the elemental composition of the watchcases and to ensure the reliability of the analytical results.

Alternatively, laser ablation ICP-MS (LA-ICP-MS) may represent an interesting alternative to ICP-MS. The advantages of LA-ICP-MS over ICP-MS notably include having comparable analytical capabilities with a much simpler solid-state sample introduction. Since no sample preparation is required, there is a lower risk of contamination, a minimal loss of material as well as a reduction of spectral interferences [132].

8.2.1 Method comparison ICP-MS vs. pXRF

The laborious sample preparation required prior to ICP-MS analysis led us to investigate the pXRF technique as an alternative for compositional analysis. Portable XRF²⁷ analysis has several advantages: it is an easy to use multi-elemental analytical technique that provides close estimates of the metal content in a very short time; the technique can be used *in situ* and is non-destructive; it is cost-effective, and no toxic reagents are needed; and it is a well-known and well-documented technique. The non-destructive nature of pXRF is particularly interesting for the analysis of counterfeit watches, since in certain cases it might be necessary to preserve the watch in its entirety. Nevertheless, pXRF has also its limitations: the geometry of the sample can greatly influence the results. If the target surface is not flat, which is the case for several watch parts, the generated X-rays could be scattered. Furthermore, due to the fact that the pXRF detection limits are much higher and that it is a surface technique, we were not able to detect the same trace metals in the watchcases (specimen set 1) and the other components (specimen set 2). It was also observed that the lower the measured concentration of a particular element, the higher the %RSD values in pXRF. Both techniques present advantages and disadvantages with regards to their attributes and analytical capabilities. An evaluation of the different characteristics can be found in Table 53. Although numerous advantages seem to stand out for pXRF analysis, its fundamental problem is of a different nature. In the presence of a surface layer, it is evident that a surface technique would not reflect the inner elemental composition. Furthermore, it is important to perform the measurements on flat surfaces, which is practically impossible for a watchcase. Portable XRF analysis was conducted without any prior sample preparation. For this reason, any differences in plating might play an important factor even though the base material could be the same for two different watchcases (e.g. the same brand and model of a watch with silver- and gold-plated

²⁷ It has to be mentioned that the initial bench-top XGT-7000 micro XRF-analyser (Horiba, Kyoto, Japan) results obtained for *specimen set 1* were not used for this research and are not further discussed. Analysis was performed without a certified reference material control, trace elements were not reliably detected and analytical problems, such as ghost peaks, arose. Nevertheless, the results were useful to identify the type of alloy, which also helped to exclude M01 from further sample preparation for ICP-MS analysis.

versions). Furthermore, pXRF does not allow measurements in the low ppm/high ppb range [133].

| Characteristic | <i>pXRF</i> | <i>ICP-MS</i> |
|-----------------------|-------------|---------------|
| Sample preparation | + | - |
| Simplicity | + | + |
| Availability | + | - |
| Cost-effectiveness | + | - |
| Destructiveness | + | - |
| Use of toxic reagents | + | - |
| Field portable | + | - |
| Specificity | + | + |
| Detection limits | - | + |
| Repeatability | + | + |
| Accuracy | + | + |

Table 53: Comparison of (positive +, and negative -) practical and analytical characteristics of the pXRF and ICP-MS analytical techniques in this research

In order to compare the ICP-MS results with the pXRF results, the same target elements (Cr, Fe and Ni) were extracted. The determined correlation values were relatively poor and the element by element paired t-test showed significant differences for all elements.

However, it should not be concluded that the two elemental analysis techniques do not agree, since the measurements were not taken at the same place. Instead, the results simply reflect that the two techniques measured two different elemental compositions. This inevitably leads to the conclusion that any further exploration of these results needs to be considered independently, as it pertains to the fundamental question of what the profile should represent. In our case, the aim was to profile watchcases based on the elemental composition of their raw material in order to gather information on their production.

The first research hypothesis for the pXRF results is nevertheless not refuted, since the obtained results appear to be reliable. Nonetheless, **we conclude that pXRF is not a suitable technique for the determination of trace elements in watchcases, especially if the surface layer is excluded from analysis.**

8.3 Chemical profiling of watchcases

The discussion in this section is based on the profiling results for the trace metals constructed from the ICP-MS results. Analysis of data obtained using pXRF can be found in section 8.3.1, which focuses on the comparison of the chemical profiling results from the ICP-MS and pXRF data, respectively.

In order to proceed, the focus should first be placed on the broader goals of a particular profiling approach. Since we sought to link watchcases from common a source and to discriminate between watchcases from different sources, the goal was to propose a method with discriminative ability. Hence, intra- and inter-source variations needed to be investigated [113].

The first aspect to clarify is defining what the intra- and inter-source variations should measure and reflect in our case. This question is, again, strongly related to the specimen set that was chosen and to where it stands in relation to the market of counterfeit watches. As previously outlined, the size and nature of the specimen set was intentionally chosen for a variety of reasons. The first restriction that was put in place was the brand. Amongst the watches produced by the chosen brand, we considered only seven models. Within the described limitations, the constructed groups are certainly valid and informative; however, the derived conclusions can neither be extrapolated to the entire market nor to other brands and models.

One of the main challenges was that we had no access to seizures with a formal indication of a common source. In this sense, the use of watches confiscated by the customs authorities was unfavourable, since they had already gone through the complete process of production, assembly, transportation and sale. Hence, any potential information of a common source becomes associated with increasing uncertainty at each step in the process. Nevertheless, it is more likely that the same raw materials were used in watches that were seized over a short period of time, which is the case for our specimen set. However, a common watchcase production source is not necessarily correlated to the time of confiscation.

In this work, we proposed to approximate intra-source variations by considering specimen- and seizure-intravariability. The specimen and seizure intravariability had comparable distribution curves. This demonstrates that watches considered to originate from a common source, based on their physical and seizure-related information,

displayed a lot of similarities between their chemical profiles. Thus, specimen-intravariability can be a reasonable approach to handle intra-source variations in watchcases with a common origin. On these grounds, our strategy was to set threshold values for each scenario at the maximum distance computed for specimen intravariability. Nevertheless, the unknown ‘true’ production-intravariability may be larger.

We found two watch pairs (M30/M31 and M28/M29), which were initially considered to measure seizure intravariability, that were subsequently qualified as outliers based on 3 out of the 4 proposed variable set scenarios. These 3 scenarios included the most abundant elements within the trace metals; hence, they are more reliable than those measured at even lower concentrations and constitute important profiling variables. Nonetheless, it cannot be excluded that these assumptions were erroneous. This inevitably raises the question of the representativeness of the considered intervariability. In order to measure the intervariability between watchcases from different production lines, the specimens that were used were those with no apparent physical similarities or relations based on their seizure. We also had no access to specimens with a formal indication of a different production source, which directly relates to the previous question on the assessment of the intravariability distribution. Nevertheless, it is worth mentioning that the intervariability distribution curves cover a broad range of distance values and is more dispersed and flattened compared to the specimen-intravariability. This indicates the possibility to differentiate watchcases based on their chemical profile. The classification performance also indicates the potential to reliably group new specimens of watchcases. These performance values are valid for the distribution curves and hence for the underlying hypothesis that the reference data used to compute these distribution curves is representative.

The chemical analysis and the subsequent profiling part of this research were done in an exploratory context. Unfortunately, we were unable to verify the hypothesis that the reference data used to measure the intra- and inter-source variations is representative. However, it was possible to demonstrate the capacity to observe similarities and differences in the chemical composition of the counterfeit watchcases. This allows us to say that the results reported in chapter 5 support the second research

hypothesis, which states that: **the analytical results allow for the grouping of specimens into classes according to their chemical resemblance.**

The information gained through chemical profiling revealed several links between the specimens, through which it was possible to group the specimen set into 15 chemical classes. Despite the small number of specimens, the number of chemical classes was high. A possible reason for this is the existence of multiple production sites responsible for the manufacturing of watchcases. Another possible explanation is that there was a large diversity in the metallic raw materials used for manufacturing these watchcases, depending on their availability.

The high number of chemical classes is less surprising considering that the brand is one of the most popular in the luxury watch segment. This leads to it generally being one of the primary targets for counterfeiting and may account for the existence of multiple production sites for the watchcases. Within the 15 groups, it was found that they generally contain at most 2 watch models, except for one group which had 3 models. This is in line with the findings of extended statistical analysis by the FH, which found that counterfeiters mainly target two models of a brand [45]. Additionally, it was determined that watches with apparently different surface plating were chemically linked. This suggests that the same raw materials are used for watches with different surface plating. Hence, our experimental results confirm the utility of analysing the inner composition of the alloy rather than using surface analysis techniques, such as XRF.

In summary, chemical profiling, in terms of data treatment and structuration of the analytical results, revealed several links between watches. Our approach exhibited promising capabilities in terms of population separation. These findings support the third hypothesis, which states that: **intelligence drawn from chemical profiling provides new insights on the production level of watchcases.** This motivates future work to validate the approach for the chemical profiling of counterfeit watches on a larger scale.

8.3.1 Profiling comparison ICP-MS vs. pXRF

The same chemical profiling approach, in terms of data treatment and structuration of the analytical results, was followed for the ICP-MS and pXRF results. It was previously determined in section 8.2.1 that the two methods are incomparable from an analytical

point of view. Nonetheless, since pXRF offers a multitude of advantages, the potential use of the data for pre-classification has to be explored. The specimen-intravariability and intervariability distributions using the pXRF results indicated the possibility to distinguish between these two populations. However, the PCA and cluster analysis revealed very different results compared to those from the analysis of the ICP-MS results. The number of chemically distinct groups was as high as that found during the exploitation of the ICP-MS results. Moreover, even a primary classification by comparing the threefold division of the dendrogram revealed significant differences. It is impossible to know what proportion of the obtained results can be attributed to the surface layer and to the underlying elemental composition. In addition, we also know too little on the plating process which further limits data comparability and interpretation. These considerations lead to the conclusion **that the pXRF results are neither suitable for use in chemical profiling in our research context nor for useful pre-classification.**

8.4 Presence of potentially toxic heavy metals

The results obtained for *specimen set 2* were interpreted with consideration of the current European Union and Swiss legislation (as described in section 2.3.2) on the presence of heavy metals in jewellery (including wristwatches).

With regards to the presence of lead (Pb), 3 of the 35 specimens that were studied showed values clearly above the legal limit of 0.05% by weight. These specimens all belonged to the low-quality counterfeit category. Visually, these watches appeared cheap,²⁸ the metal was occasionally stained or dirty and the watches were very light. Although the size of the specimen set considered in this study was very limited ($n=35$), the fact that 3 out of the 12 low-quality specimens exceeded the Pb limit is alarming. Since the watches in the two other categories were within the outlined limits, the results also suggest that for the higher quality counterfeits, there is a lower chance of finding specimens with potentially harmful elemental contaminations.

Our analysis of the imitation gemstones revealed very high concentrations of lead in two specimens. Fake gemstones can be made from a variety of materials, such as different types of glasses and plastic, and crystal glass typically contains lead. In the REACH legislation, crystal glass falls within the exceptions for the application of lead regulations, due to a lack of suitable alternatives at that time. In the future this is likely to change, since alternatives currently exist; for example, Swarovski introduced their lead-free crystal formulation in 2012²⁹.

It was also observed that these imitation gemstones are not tightly adhered to the surface; some stones were already missing on acquisition and others fell off during routine handling for analysis. Because of the risk of accidental ingestion, it is clear that counterfeit watches that contain crystal beads constitute a significant risk for children. Cadmium (Cd) was not detected in any of the specimens; however, considering the limited specimen size ($n=35$), this finding does not mean that cadmium is not present in other counterfeit watches.

²⁸ i.e. non-experts could easily recognize these watches as being counterfeits.

²⁹ Information retrieved from the official Swarovski website: https://www.swarovskigroup.com/S/aboutus/CSR-Product_Safety.pdf (last accessed: 02.03.2020)

Portable XRF also demonstrated its limitations as a screening technique for the determination of toxic heavy metals. We observed that different measurements taken on the same watch part displayed considerable variances. In addition, correct sample placement on the benchtop stand is challenging and it is difficult to know the exact position at which the measurement was taken. This is partly due to the relatively large measurement window, but also due to the different shapes of the watch parts. The measurements of the fake gemstones were particularly challenging, since they are very small and adhere to the metallic surface which affects the results. Thus, it is essential to consider the pXRF results as preliminary information that needs to be confirmed by other studies.

The presence of Nickel (Ni) is regulated by the quantity that may be released from jewellery and other products that are intended to come into direct and prolonged contact with the skin and is defined as a migration limit. The regulations were established to reduce nickel in consumer goods, since almost 10% of the general population are believed to have an allergy to nickel [134]. Nickel is used as an alloying element to increase the corrosion resistance and offer better malleability³⁰. Due to the crystal structure of stainless steel, the nickel that is present is not free to react and does not constitute a problem in terms of leaching. Nevertheless, in alloys other than steel, leached nickel can be a problem. The strip tests we conducted for nickel release indicated positive results on several parts of the counterfeit watch specimens. However, the test clearly provides very little information with regards to the actual quantity of release and whether it is below or above the legal limit. Nonetheless, the test indicates whether nickel is released and is especially useful since a high nickel content in a matrix is not necessarily correlated to its release from the matrix. This test is also an indication for allergy sufferers and is commercially available in pharmacies. It should be kept in mind that nickel could also be released from other parts that have a nickel content below 10%, but not all parts were systematically tested. Interestingly, the positive nickel test specimens also all belonged to the group of low-quality counterfeits (5 specimens out of 12), except for M20 which belonged to the average quality category.

³⁰ Information retrieved from the Steel Glossary of the American Iron and Steel Institute:
<http://www.steel.org/steel-technology/glossary/glossary-a-e.aspx> (last accessed: 02.03.2020)

The higher quality counterfeit watches were more frequently made of steel and had a lower risk of nickel liberation.

For a more comprehensive risk assessment, further testing on a larger specimen set including low quality counterfeits is necessary. Portable XRF could readily function as an initial screening technique. However, due to the previously mentioned limitations, we recommend using other analytical methods in conjunction for confirmation, as suggested by the standard operating procedure for the determination of total lead in metal products issued by the ‘Consumer Product Safety Commission’ (CPSC) [111].

To monitor nickel liberation, the test strips could be used as a rapid and cheap qualitative test, but it would certainly not be suitable to determine compliance with legal requirements. However, selected specimens could be submitted to the reference method for the release of nickel (EN1811:2011 + A1:2015) [135]. This method involves soaking the item in an artificial sweat solution and analysing the quantity of nickel dissolved in the solution.

During this work it was possible to gather information on potentially harmful elements from a very small specimen set of counterfeit watches using simple testing methods. The indication of lead and nickel contaminations present in low-quality counterfeit watches was alarming, demonstrating that counterfeiters do not comply with safety standards for the alloys that they use.

The above-mentioned results hence support the fourth research hypothesis which proposed that counterfeit watchcases contain potentially toxic heavy metals.

Using the outlined methods, we were able to demonstrate that some counterfeit watches are manufactured with inferior materials. Nevertheless, it is also necessary to test whether the surface coatings fail to protect consumers from chemical harm arising from contact with the lead content of the base-metal watchcases. In addition, paints and dyes used in the watch should also be tested to find out whether they contain unsafe levels of toxic compounds.

A report issued by the OECD on the economic impact of counterfeiting and piracy stressed the necessity to document the adverse effects that counterfeit goods can have on public health and safety [11]. Our results also present a more comprehensive understanding on the effects of counterfeiting; demonstrating that they are not just

restricted to intellectual property and trade infringements but may also pose a public health problem [136].

8.5 Physical profiling

The intelligence gathered from the meticulous analysis of all imprints present on the watches of *specimen set 1* and their links with formerly analysed watches are discussed in this section. **The following findings support the fifth research hypothesis, which stated that: a thorough study of all imprints found on the specimens, as well as their relationships with previously analysed watches creates new intelligence and insights.** This allows for a deeper comprehension on the use and scope of the tagging tools and on the structures of the underlying assembly sites.

8.5.1 Physical links between the 35 watches

For this research, the main descriptor of a counterfeit watch on a physical level was the data set containing the codes which described all present imprints. The methodology applied by the FH includes analysing each watch component separately; the case including the back, the strap including the clasp, the crown, and the dial. A visual representation of the links between the different components clearly revealed that these links have to be considered separately for each part. Some watches shared similar imprints over their entire body; however, several watches were linked through imprints on only one component, with no evident links on the rest of the components. This indicates that the tagging of watches did not take place in a single step, but rather that the different parts were tagged individually and then assembled to form the entire watch.

This is particularly true for the imprints found on the crowns and on the clasps (brand logo application). The observed physical links between the 35 watches suggested that these imprints generated several connections between specimens that were otherwise not related. This indicates that the crowns and application of the brand logo on the clasps were produced independently and then distributed to the different assembly sites.

For the dial, links were only found between watches of the same model. This was not surprising since the dials are generally where the model is displayed, hence these imprints are for the most part model-specific. The imprints are useful for distinguishing between different production lines for counterfeit watches of the same model, but less

so in terms of identifying connections to large-scale production lines of counterfeit watches.

The physical links between the different parts of the watches are fairly complex and it was difficult to form an understandable structure. Overall, numerous links exist, and most of the specimens were connected in some way. Only specimens M02 and M08, which were linked to each other through the imprints on their crowns, were observed to be completely separated from the other 33 watches.

The interconnection between all watch components suggest that the production of these watches is the result of an interconnected network that interacts on several different levels.

It was found that limited number of codes was responsible for a large number of links, since 145 of the codes were not linked within the specimen set. These codes may still be connected to other watches that were previously analysed. Hence, it is important to analyse imprints in a more extensive context, which is discussed in the next section.

8.5.2 Physical links on extended level

To more extensively analyse the imprints, the number of individual codes with links increased to 190 and links with 504 other watches were found. These findings indicate that the considered brand and models of watches were counterfeited on a large scale within connected networks, and it was possible to obtain a great amount of new information based on an extensive analysis of imprints from the initial 35 watches.

For the extended study of the imprints, it was very useful to prioritize imprints that generated numerous links with previously analysed watches as well as their classification into brand-, model- and non-specific imprints.

It is necessary to perform statistical evaluations with regards to the brands and models most likely to be counterfeited. In addition, it is also important to consider the imprints that generate the most links. In total, 27 imprints that generated links to at least 25 other previously analysed watches were examined more closely, from which valuable information could be retrieved. Considering only these 27 codes, instead of the initial 190, more than half the number of occurrences in the database were found, as well as nearly three quarters of all links to other previously analysed watches. A special

emphasis was placed on these codes since they suggested an industrial-scale application of the tools that were used to produce them.

Four different groups of codes, with each group formed by codes generally found in combination on the watches, were identified. This suggested that the corresponding imprints were tagged at the same time. There was no mixing between the groups, meaning that codes of different groups were never found on the same watch. All the corresponding imprints were located on the straps and indicated four different production lines for the straps.

To narrow the field down, we decided to focus on codes generating at least 25 links, nevertheless, a large number of codes (163) generated between 1 and 24 links to previously analysed watches. Thus, it is up to the analyst to decide which codes should come under further scrutiny. The fact that, in our case, the selected codes generated more than three quarters of all links demonstrates that it was a well-reasoned choice. Furthermore, 3 imprints found on the crown could link 111 watches to each other, indicating 3 major production lines for these crowns. Generally, the imprints on the clasps and crowns generated a considerable number of links. This reinforced the hypothesis that they had been produced separately on a large scale and were then distributed towards multiple assembly sites.

For analysis of the watchcase, one single model-specific code generated links between 45 watches. All other codes from the watchcases generated less than 25 links within the database. None of the imprints found on the dials linked to more than 25 watches, even though links with other watches existed. The reason for this could be that the imprints on the dial are mostly model-specific; they generally link counterfeit watches of the same brand and model that have been tagged with the same tool. Hence, the dial should not be considered as the component of interest if the goal is to look at the imprints with the most links, which is indicative of an industrial production. Nevertheless, these imprints could be informative to study relationships between watches of the same brand and model.

Among all the imprints, there were 145 codes that did not generate any connection within the 35 watches. However, after analysing all the codes within the entire database, only 38 were not linked to any other previously analysed watch. It is nonetheless important to consider these imprints, even though the tools generating

them were rarely used and not likely to reveal production on an industrial scale. However, care should be taken to avoid missing links between the initial specimen set. Within this set of codes, links between the watches of the specimen set were found, meaning that these codes were analysed for the first time. It was found that most of these imprints are on the case and on the dial and almost half of these codes were also model-specific.

The gathered information on all the uncovered links could have an important role if an assembly site is dismantled. If tagging tools were to be found, it would be relevant to check whether they correspond to imprints found in the database and to how many watches they are linked. This may be used as additional evidence to support the fact that an assembly site produced replica watches on an industrial scale.

8.5.3 Temporal analysis of imprints and geographical spread

All the seizure-related expert reports contained information related to the country of seizure, as well as the date on which the analysis of the counterfeit watch by the FH began.

For temporal analysis of the imprints, only codes that were found on at least 25 other counterfeit watches in the database were considered. All 504 watches were used for retrieval of information related to the geographical distribution, which means all watches that were linked via at least one characteristic to the 35 watches of the original specimen set were used. Temporal and geographical information in this context are interesting but it is important to understand what the data represents and how it can be interpreted to produce intelligence.

Furthermore, ten years after the first seizure of watches containing these imprints, new watches with the same physical characteristics reappeared on the market. The temporal persistence of imprints clearly revealed that the tools used to produce them are used over a long period time. It should be noted that the complete analysis was based on the initial 35 counterfeit watches. The way the temporal analysis was performed can only reveal at what time a specific code was identified for the first time, and when and how many times it was found up till the point that the watch of the initial specimen set was analysed. As a consequence, it is not possible to detect whether a certain tool, for whatever reason, is no longer in use. The fact that some codes exhibited the same

temporal profiles strongly supported the hypothesis that they had been used by the same manufacturer.

Watches that were linked through physical profiling had been seized in 13 countries on three continents. This suggests that the studied counterfeits were produced on a regional scale, which is supported by the multiple relationships in their physical links, and then sold globally.

8.5.4 Recommendations for database management for FH

The systematic analysis of imprints and their introduction into a relational database began in 1996 at the anticounterfeiting department of the FH [137]. Hence, a well-structured and extensive archive of traces and information already exists. Currently, an expert report on a specific case contains the identification of all imprints and provides all identified links to previously analysed imprints and their corresponding watches. This approach is nevertheless mainly based on isolated cases, and the knowledge that the report represents in relation to current problems, patterns, series and linked cases is missing.

The database has a great potential to gather more information and produce intelligence on watch counterfeiting; more precisely, to understand the scope of the illicit activity at identified assembly sites, their volume of production, period of activity as well as geographical information on the origin, provenance and country of seizure of counterfeit watches. An effective forensic intelligence process would advise the logical processing of newly obtained information and its integration into the database that contains previously acquired information. This follow-up process is dynamic and iterative, with the aim of providing up-to-date exploitable knowledge on the phenomenon under consideration.

From the above-mentioned findings in section 8.5, several recommendations for improved monitoring of serial phenomena in the database can be made. These recommendations reflect a perspective to detect, analyse and prioritize imprints that are the most often used and that indicate an industrial-scale production of counterfeit watches, which may not always be the question of interest. On average, the FH conducts detailed technical analyses on 600 watches per year (2011-2017) [45, 47, 138-141]. The meticulous physical analysis of each timepiece is very time consuming. The

following suggestions are thus formulated with the view of optimizing the organisational performance, in terms of reducing the work expended on each watch, and simultaneously gathering the maximum amount of information that can be used in an intelligence perspective. All proposed improvements are solely based on the findings that emerged from the in-depth analysis of the imprints present on the watches in our specimen set. However, we can assume that the market for replica watches of other brands may be organised in a similar way and that the proposals could be applicable to a far broader context.

Broadly speaking, imprints that are model-specific and imprints located on the dials and cases revealed the least number of links between the considered watches. It may be beneficial to instead prioritize imprints on the strap and on the crowns, while the imprints on cases and especially on the dials could potentially even be omitted. The same applies to model-specific imprints, in fact the vast majority of model-specific imprints can be found on the dial. Furthermore, the clasps should constitute a separate category, since they do not appear to be related to the other imprints observed on the strap.

A possible approach for testing if such measures prove to be adequate and useful is conducting an in-depth database analysis. First the target brands and models should be prioritized. For this, the results of analysis performed by the FH identifying the statistically most counterfeited brands and their respective models can be used [47]. Initially, it may be useful to consider a selection of such commonly counterfeit brands separately. Codes that generate the most links within the specimens of the considered brand should then be identified. For this, a similar procedure as proposed in Chapter 6 could be applied, however all specimens of a specific brand should be considered. It is up to the analyst to decide what a large number of links signifies and what number of links shall be considered for further exploitation, with consideration of the initial number of specimens used.

For the purpose of this research it was demonstrated that it is useful to designate imprints in relation to their brand- or model-specificity or if they are non-specific. Hence, ‘specificity’ could be added as a further attribute of the imprint entity in the relational database. The data can thus be extracted, filtered and processed according to watch components and specificity. Furthermore, temporal analysis of the imprints,

as well as country of origin, provenance and seizure of the watches should be included. It is conceivable to assign different priorities to the imprints based on the number of links they generate. Subsequently, the higher priority ones could be followed more closely, allowing for a more immediate detection of new cases that are related to an active series. It would also be beneficial to identify a group of imprints that are (often but not necessarily always) found together and defining them as a class. Such imprints are logically found on the same watch component and strongly indicate that they were produced at the same time and the same place.

Imprints that do not generate links (over a long period of time) indicate that the tools that generated them are no longer or rarely used and are not likely to reveal production on an industrial scale.

Since the FH database already consists of several thousand records and its size and complexity continue to grow, an obstacle referred to as ‘linkage blindness’ [142] may occur. Linkage blindness can be defined as “*organizational or investigative failure to recognize a common pattern shared on different cases*” [53]. Here, a network of links may be difficult to identify and no longer provide a clear overview. This can be countered by identifying a promising set of cases, which requires a limited number of specimens that are selected as proposed above by proceeding in a brand by brand manner for example.

8.6 Contextualisation

A counterfeit watch constitutes physical evidence of an illicit activity and contains physical and chemical data on multiple levels, as well as spatiotemporal information related to the seizure and indications on the source of fabrication. The combination of these different sources of data is a crucial step towards a deeper comprehension of the structure of the criminal organisations. Link analysis was performed and visualizations combining both chemical and physical connections were produced with the goal of deciphering specific patterns in the data. This in turn allowed us to formalize propositions about the production and distribution networks for the counterfeit watches.

Numerous chemical classes of watchcases were identified by profiling according to their elemental composition. It was observed that several watches had no further links to any other watches and displayed single chemical classes. Two propositions could be made according to these findings. The first possible explanation is the presence of numerous production sites that are specialized in the manufacturing of watchcases for the studied brand. However, there is the possibility that a production site procures its supply of metallic feedstock opportunistically from different sources. In this case, the chemical profiling would also lead to the identification of distinct chemical classes, even though the watchcases originate from the same production site. Inevitably, this prompts the question of the representativeness of the elemental composition as a reflection of the production process. In this case, the chemical links represent the alloy production process but not strictly the watchcase production. This indirect relation leads to interpretation difficulties and points to the importance of gathering further information on how counterfeiters access raw materials. This in turn would enable the contextual interpretation of the analytical results to actively transform the data into actionable intelligence.

Overall, a low level of correspondence between the chemical and physical links was observed. Demonstrating that the chemical profiling added complementary knowledge that was previously unknown. Furthermore, these links occur at the production level, prior to the tagging of imprints on the different components. This finding supports the hypothesis that the production of the watchcases and the tagging of watch components take place at different stages. Additionally, the watchcases could be produced at several

different production sites, as supported by the numerous chemical classes, and then followed by distribution towards multiple, independent production sites.

A number of chemical links were corroborated by the physical ones. This was mostly the case for watches of the same model that were visually indistinguishable and had the same seizure context. For these watches, chances are high that they originated from the same production and assembly line.

On the other hand, both chemically linked watches without physical links and physically linked watches without chemical links were found. This supports the hypothesis of a large-scale connected network and is further underpinned by the comparison between the cases and the imprints on the cases.

It was hypothesized that several watchcase producers supply multiple assembly sites that possess tools for tagging the cases, straps, and dials. This is supported by the numerous physical links that exists between these components. The crowns and the logo used for the clasps seem to be produced on a large scale with a few producers supplying several assembly sites. This proposition is reinforced by the fact that these imprints generated multiple links between watches that were previously neither physically nor chemically linked.

As an interesting example, it was found that three replica watches of the same model, with the same colour and no major visual differences, nevertheless, belonged to three different chemical groups. Among the 14 imprints on each watch, there was a direct correspondence for 9 imprints between the three specimens. The remaining imprints were of the exact same type but displayed differences that suggested that they were produced using different tools. It was possible to superimpose the imprints if they consisted of several lines or had a large spacing between two words. It is possible that some imprints that are considered to be made of a single tool are in fact the product of multiple tools; therefore, more links between imprints could exist. However, the superimpositions were made without any reference measurements. It may be beneficial to search the database to verify if such possible misclassifications exist on a significant scale. If this were the case, imprints consisting of several lines or spaced portions (such as SWISS MADE) should be analysed separately. Differences may also be found due to distortions that resulted during the tagging of imprints, especially on components where the surface is not entirely flat. Furthermore, an assembly site may also have

several tools for the same imprints at its disposal, which results in different physical traces for the same assembly site. Nevertheless, our findings indicate that multiple links can be found on the basis of these physical traces.

Chemical links represent a single variable, namely the chemical class; hence, watches can either be linked or not. Furthermore, these chemical classes refer only to watchcases. The situation is different for physical links between two watches. These links can occur between the totality of the imprints, between some of them or none. This information also has to be interpreted, allowing us to derive evidence on whether parts were tagged together or separately. Hence, chemically profiling other watch components may provide different results.

Over the course of this work, every source of information (chemical, physical or spatiotemporal) provided unique new insights and added to the existing body of knowledge. Although these findings cannot be extrapolated to the production and distribution of the entire counterfeiting market, we were able to provide valuable methods and a solid basis for further research. This tends to confirm the sixth and final research hypothesis which stated that: **the comparison between the chemical and physical profiles provide valuable intelligence on the distribution of counterfeit watches towards the assembly sites, resulting in a deeper comprehension of the relations between these two steps.**

8.7 Global discussion

An important initial component of this thesis was the detailed review of literature on counterfeiting and, more specifically, on counterfeit luxury fashion goods that include replica watches. Reports on counterfeiting written by governments and various institutions were studied. These reports mainly included information on the magnitude (number of cases and articles) and scope (product categories) of identified counterfeit goods; their provenance and transport; and estimates of their economic impact and resulting adverse consequences. However, a direct measurement of the counterfeit market is impossible using current methodologies, and it must be accepted that the true magnitude remains unknown. Nevertheless, these reports reflect an awareness of the problem and demonstrate the global nature of the counterfeiting phenomenon, justifying its investigation from a forensic research point of view.

While research on consumer attitudes towards counterfeiting is abundant, it is nonetheless difficult to prevent consumers from buying counterfeit goods, criminalize its possession and prove that buyers knowingly purchased them. Public awareness campaigns are of great interest to reduce the demand for counterfeit goods.

From a supply-side perspective, the data pool of available information clearly needs to be strengthened. All parties with an interest in anticounterfeiting, which include governments, businesses and research institutions, should work to improve the collection of information and, in particular, its analysis. The timely collection of relevant information and the detection of actionable insights are essential for the design and implementation of effective countermeasures. Counterfeiting is not an area of research on its own, but rather a multi-disciplinary gathering of information. However, our existing body of knowledge only partially reflects its complexity and still lacks an understanding of the mechanisms and structure of this illicit market. Forensic research, and in particular, forensic intelligence on counterfeiting is nearly nonexistent, although some excellent initial work has been carried out in various fields. The primary reason for this is that counterfeiting, especially of luxury goods, is not perceived as a security threat in society, and hence, is a low-priority in terms of law-enforcement.

The fundamental idea of forensic intelligence is to use acquired traces to produce intelligence on the phenomenon under investigation. Even though this general approach

is not new, extending it to new areas of interest inevitably raises new questions and challenges that need to be addressed.

In order to conduct forensic research on the counterfeit watch market, many choices have to be made that ultimately affect the intelligence that can be gained. Thus, it is the role of the forensic scientist to identify and characterize the nature of the material traces and whether they are likely to predict a common source based on their physical and chemical features. The key question is nonetheless the formation of a clear definition and understanding of what these traces reflect. How can the traces explain what occurred and at which level³¹ of the counterfeiting process can they provide information? By using the physical object from the illicit activity as a starting point, various characteristics can be measured. We currently have a vast number of possibilities for analysis at our disposal and the ability to perform a range of physical and chemical measurements. Given the range of experimental possibilities, an extensive literature research (not only academic) and discussions with experts on watch counterfeiting was an important initial step to formulate the research questions. In the subsequent step, it was then possible to identify the material traces with the potential to respond to some of these questions.

Through this preparatory process, we determined that there was a need to gather more information on the production of individual components; to study the structure and organisation of their subsequent distribution towards assembly sites; and finally, to identify how these steps are interconnected.

For the production level, it was decided that the elemental composition of the watchcases would be used as a marker of their origin. Since CNC machines are used to cut out cases from solid pieces of metal [103]; it is reasonable to assume that only a finite number of production sites that intend to manufacture copies of watch parts have access to such technology. The fabrication of watchcases represents a key step in the production process. This is because their unique shapes may be more characteristic of the brand and model that was intended to be counterfeited than the other components. The other components, such as the straps, could possibly be acquired from legal production chains. Nevertheless, the chemical and physical analysis of the other watch

³¹ The term 'level' refers to the three different steps of production, distribution and diffusion of counterfeit watches.

components might lead to very different results. While the different components may have been produced individually in legitimate factories, they nonetheless have to be assembled together at some point. Consequently, the assembly of a watch is a further key step. It is believed that the tagging of imprints takes place at the assembly stage. The use of trademarks and indications of source are legally protected and the line between legal and illegal activity, in terms of intellectual property rights, is crossed at this step. The traces left from this process, namely the imprints, are highly informative with respect to the structure, scope and activity of the assembly sites.

At this point it is useful to draw comparisons between chemical and physical profiling. From the start it seems clear that this comparison is difficult and also unequal at present; there is clearly much more information that could be gathered from the physical analysis of imprints. Furthermore, it needs to be underlined that the systematic recording and analysis of imprints present on counterfeit watches by the FH has been in progress for more than 20 years. The database is constantly filled, maintained and a significant amount of detailed information is available.

It should be emphasized that the sample preparation and ICP-MS analysis for the watchcases was especially challenging at the beginning. While the final contribution to the understanding of the production step may seem limited, a lot of effort was put into the development of the analytical approach. We successfully managed to develop an analytical approach to both reliably qualify and quantify trace metals in steel cut samples and to assess the possibility to discriminate between trace metal profiles. This analytical strategy may also find applications in other fields of research where the elemental composition of steel products is of interest. However, trace metal profiling requires a significant allocation of resources and the methodology needs to be tested on a larger specimen set. Sets used in further studies also imperatively need to contain specimens known to originate from common and different sources in order to correctly evaluate and validate the method. Generally, the aim of a profiling method is to maximize the content of information by granting priority to methods that are the least resource-intensive and destructive. Hence, it is fair to say that profiling through classification of the physical characteristics of the imprints currently offers several advantages. However, profiling based on just the visual characteristics does not appear to be a viable alternative; it was possible to observe watches that were visually very

different that could nonetheless be grouped based on their chemistry or linked via their imprints (even for watches of different models).

Considering the physical links and the associated imprints of the 35 watches with the entire memory of the FH database, more than 500 other counterfeit watches were connected with our research set. These findings displayed the possibility to achieve even greater insights into these interconnected networks. Furthermore, this proceeding allowed us to identify the types of imprints (as well as their location on the different watch parts) that generated the most links. These imprints are particularly interesting since they suggest an industrial use of the tools that were used to produce them.

Throughout the course of this work, no special emphasis was given to the studied brand (though some brand-specific aspects of the imprints were considered); hence, the process can also be generalised to other brands analysed by the FH.

Forensic intelligence fundamentally depends on features of traces that can be measured, compared and compared; in addition, this information is combined with other forms of information on the phenomenon of interest. We were able to demonstrate that new, previously unknown information could be gathered from the chemical analysis of watchcases. Furthermore, the in-depth study of the imprints and their relationships can push the interpretation on distribution patterns of counterfeit watches further. Although based on a limited number of seized specimens, our research revealed that the production and distribution of counterfeit watches is highly organised. This leads to the hypothesis of a main organised network, with many producers and assembly sites that acquire different watch components separately. The number of links found with previously analysed watches suggest an industrial scale production of watches, with links stretching back over a period of 10 years in some cases. Although an organised structure for the production and distribution of counterfeit watches had been assumed, the results of this work further corroborates this hypothesis, at least for the watches in our specimen set.

At this point it is important to critically discuss the contribution of the obtained results in view of implementing investigative or preventive measures against watch counterfeiting. As mentioned above, the physical profiling of imprints offers several advantages in terms of resources and, at the same time, offers a very rich source of information. Hence, it is also a pertinent choice for the systematic analysis of

counterfeit watches. Nevertheless, on the basis of this work, several suggestions for improvement were made. These particularly refer to the monitoring of the database to gather more information and to produce intelligence on the market. Possible options include the prioritization and selection of imprints which are the most widely used. Furthermore, as the size and complexity continue to grow it might be difficult to identify networks of links and to maintain an overview. We proposed to identify a promising set of cases and to analyse the physical features under the consideration of the above-mentioned recommendations. Circumstantial information combined to imprint profiling allows for a first overview on the relationships that may exist between watches of a specific specimen set. We recognize that the developed chemical profiling method will, at least at this stage, not find a systemic application in the current anticounterfeiting process. However, a systematic implementation must not necessarily be the prime objective. Chemical profiling may help adding a chapter to the story. In some cases, it might be useful to further study and validate existing hypotheses emitted during the physical analysis. For example, it might be interesting to find out whether watches that exhibit superficial similarities were made with the same base alloy and hence likely to originate from the same fabrication site. Depending on the results, different decisions or operations may be initiated. In this sense, the process can be seen as an intelligence-led process. In general lines, this is consistent with the proceeding during this research, although our research was initially focused on the development of a chemical analysis and profiling method. The specimen set was based on a relevant selection of counterfeit watches and we tried to gather a maximum of information about their fabrication and distribution.

Elaborated chemical analysis may also be very useful in the future when technical analyses approach their limits. In fact, quality improvements observed in 2018 relate to all watch components, commercial documents and delivery methods [143]. The technical capacities of counterfeiters progress constantly, and it might be necessary to conduct more and more specific analysis to determine whether a particular specimen is indeed a counterfeit or not. The proposed trace metal profiling method also provides a solid groundwork in this perspective.

Our work demonstrates the potential of material traces to provide significant insights into the counterfeiting networks that they originate from. Watch counterfeiting is

obviously not limited to their fabrication, hence, aspects related to the subsequent steps in the supply chain are also of great interest. Forensic intelligence is a wide area of research and takes on a variety of forms. It is certainly only part of the solution and has to be combined with other sources of information and interpreted accordingly [58]. We are nonetheless convinced that it is crucial to tackle the problem at its source and to obtain an appropriate understanding of the structure of the network involved in the initial fabrication of counterfeit watches.

In this sense, collaboration between research institutions and those interested in the illicit events is necessary to successfully evaluate and validate the potential practical implementation of the developed methods. Over the course of this work, we were able to uncover new information that led to a deeper understanding of certain aspects of the counterfeiting process, and to propose several recommendations to optimize the use of existing information. This research exemplifies an essential part of the broad definition of forensic science as “*the utility of the sciences as they pertain to legal matters*” [53]. The issues that forensic scientists face concerning material traces are generally similar, such as their existence, use, and interpretation. In order to address these problems, a multi-disciplinary collection of contributions from domains such as analytical chemistry, statistics, watchmaking, and intellectual property rights was necessary to conduct this research. Anticounterfeiting efforts generally involve several actors that need to collectively participate in the investigative and preventative process. In this perspective, we were able to demonstrate that forensic scientists can also provide a valuable contribution.

8.8 Limitations and perspectives

In this section, the global limitations of the proposed methodology as well as future challenges and perspectives are reviewed.

Several limitations regarding the analytical strategy, the chemical profiling methodology and exploitation of the physical characteristics were already addressed throughout the discussion section. It is clear that the major limitations of this study relate to the limited number of specimens, their nature and the lack of specimens of known origins (common and different). Nevertheless, it was demonstrated that a significant amount of information could be gathered from a limited number of specimens and that the specimen set size should not necessarily be seen as a limiting factor.

It is also important to mention that the findings of this thesis were valid at the time the expert reports were issued. The methodology was based on a specimen set with seizures from 2015 and the retrospective analysis of the database that was conducted. The definition of forensic intelligence as “*the accurate, timely and useful product of logically processing (analysis of) forensic case data (information) for investigation and/or intelligence purposes*” [58] implies a current temporal component. While the situation may have evolved, the type and scope of knowledge that can be gained from the chemical, physical and spatiotemporal information remains unchanged and should be the point of focus.

The physical and chemical information are both associated with uncertainties that are not strictly at the technical level. Nonetheless, the material traces are extremely rich in information and emphasis should be placed on their contextualization, even though the information may be flawed, incomplete and fragmentary [54]. In the context of forensic intelligence, we are not searching for a ‘scientific truth’; rather, it should be seen as a process that offers valid and informative insights to reason upon in order to advance the knowledge on a particular type of illicit market.

The information that can be gathered from the study of objects that are material traces left from an illicit activity, depends first and foremost on their seizure or collection. Regrettably, the Federal Customs Administration in Switzerland instituted an austerity program including the reduction of officials assigned to verify and impound counterfeit watches, resulting in 75% less inspections and seizures in 2017. Such legal

and political changes clearly cause linkage blindness [57] by compromising the work of the FH anticounterfeiting department; and consequently, their ability to understand and explain watch counterfeiting [47].

The main specimen set used for this research consisted of watches seized by customs in Switzerland knowing that other sources could also be considered. Currently the vast majority of counterfeit watches submitted to technical analysis by the FH involves customs seizures (tourist traffic and postal packages). Other alternatives would be seizures from large scale police actions in assembly sites or warehouses or to physically purchase counterfeit watches or to order them online. Depending on the question, the suitable source of specimens has to be determined. Specimens from the same fabrication site for example would allow to determine a more representative production-intravariability, which in our case had to be approximated by other means. However, such specimens are also difficult to access. Counterfeiting has been amplified in recent years with the advent of e-commerce and social media. With regards to watch counterfeiting, it would be very interesting to combine material traces and digital data from online sales channels. Such supplementary sources of information on the diffusion of counterfeits towards online marketplaces would fill another important piece of the puzzle. This could possibly yield a more refined understanding of the activities of the supply chain networks for counterfeit watches. Furthermore, replica watches acquired from virtual sales channels may be more representative of supply and demand for this market than seizures by customs authorities.

Generally, much more research on product counterfeiting could be done from a forensic research point of view. An interesting and concrete application could be the use of forensic intelligence against counterfeit cosmetics³² (also referred to as 'fake-up'). The cosmetic industry has experienced an enormous resurgence over the past few years, especially with their promotion on social media³³.

³² The U.S Food and Drug Administration (FDA) defines cosmetics as "...articles intended to be rubbed, poured, sprinkled, or sprayed on, introduced into, or otherwise applied to the human body for cleansing, beautifying, promoting attractiveness, or altering the appearance.",

retrieved from: <https://www.fda.gov/Cosmetics/GuidanceRegulation/LawsRegulations/ucm2005209.htm> (last accessed 25.09.19)

³³ <https://cdn2.hubspot.net/hubfs/2018953/Research/RedPoints-Market-research-survey-counterfeit-cosmetics.pdf> (last accessed 02.03.2020)

Since cosmetics are products that come into direct human contact, the potential health risks are profound; this in turn is also poses a very serious threat to a brand's reputation. According to media reports³⁴, it was found that counterfeit lipsticks and eyeshadow palettes contain unsafe levels of lead and other toxic heavy metals, bacteria and even animal waste. Such products can seriously put the health of consumers at risk. Consequently, there is a strong interest for anticounterfeiting in the cosmetics industry where consumer health and the brand's reputation are at stake. Although public health assessments are outside the forensic science domain, in the course of performing chemical analyses for use as forensic case data, the analytical results can additionally be used to assess potential health and safety related risks. A profiling methodology based on the chemical and physical features (including the product and the packaging) could be developed for counterfeit cosmetics. Through a detailed study of the revealed links, new insights into illicit products and their markets could be gathered. Areas of interest would be whether such products are the fruit of an industrial-scale, organized network or rather from unrelated small-scale production facilities. This in turn could support consistent decision-making in terms of the priorities for investigative work. The presented example is only one possible application, and the general approach could be applied by analogy to other illicit market domains that face similar problems.

³⁴ <https://www.cbsnews.com/news/fake-makeup-health-hazard-us-customs-and-border-protection/>
(last accessed: 02.03.2020)

<https://www.dailysun.co.za/Umswenko-Wasesun/the-dangers-of-using-fake-make-up-20180622>
(last accessed: 02.03.2020)

<https://www.healthline.com/health-news/counterfeit-makeup-a-health-danger#1> (last accessed: 02.03.2020)

9 Conclusion

Counterfeiting is an illicit multibillion-dollar industry and remains, despite many efforts, a worldwide scourge. Contrary to the general perception of counterfeiting having minimal real-world consequences, it can indeed have serious adverse effects on the economic, social and human levels. Significantly, the profits derived from the industry are largely associated with revitalising other forms of underground economies and illicit activities with more acute consequences. By definition, counterfeiting is a serial criminal activity and a broad range of consequences can be associated with it; as such, all public and private stakeholders that have a direct and indirect interest in the anticounterfeiting process should contribute to the development of accessible and efficient means to combat this phenomenon.

Luxury goods and apparel are particularly affected and remain the preferred targets of counterfeiters. Among the luxury goods, Swiss brand watches rank among the top counterfeit products for many years and pose a serious threat to an important industry in Switzerland.

The legal- and anticounterfeiting department of the Federation of the Swiss watch industry (FH) strives to combat counterfeiting. One of their core activities is the physical examination and analysis of counterfeit watches. The approach adopted by the FH relies on linking counterfeit watches based on the codification and precise measurements of their physical features. Of particular interest are the imprints of corporate logos, brands, and technical- and geographical indications present on the different watch components. Once established, these links often provide information on the assembly site, which is the final step in the fabrication of a counterfeit watch. However, very little is known about the production of these watches and its relationship with the assembly step. Since the criminal counterfeiting networks are highly agile and dynamic, developing appropriate countermeasures against the traffic of counterfeit watches requires a broad comprehension of the phenomenon.

Forensic intelligence studies criminal activities using the investigative and logical interpretation of forensic case data, i.e. traces, to support a great variety of decision-making. Counterfeit watches contain traces at the physical and chemical level that can be related to their production and/or distribution. Since this data comes directly from

the remnants of illicit activity, studying these traces from a forensic intelligence perspective can actively contribute to fostering an improved and refined understanding of the criminal activity. In this sense, it is of profound importance to understand the level of intelligence that traces, as well as circumstantial information, may provide. For this work, in addition to a comprehensive literature research, the close collaboration with the Federation of the Swiss Watch Industry allowed us to define the research questions for this thesis. This groundwork resulted in the decision to focus on the production of individual components, to study the structure and organisation of their distribution towards assembly sites, and finally, to find out how these steps are interconnected. The proposed approach aims to apprehend criminal activities based on the trafficking of counterfeit watches, through the contextualization of information and traces. A complementary objective was to appraise the potential risk to public health due to the presence of toxic heavy metals in watchcases and its relevance as an anticounterfeiting approach.

The first practical goal was to investigate the possibility to obtain reliable data at the production level through analysis of the elemental composition of watchcases. The underlying reasoning was that watchcases originating from the same production batch are made from the same raw materials and should, therefore, have the same chemical composition. The following objectives were outlined: develop an analytical strategy to measure the elemental composition of watchcases; analyse a selection of counterfeit watches; propose a statistical methodology that discriminates between watches of different origins; group the watches according to their chemical resemblance; examine the relationships amongst them; and finally, to provide new information at the production level. The first specimen set considered for this research consisted of 35 counterfeit watches of the *Rolex* brand. The set was composed of seven different watch models from the brand and were seized at the Swiss border over a short period of time. The *Rolex* brand was chosen because it is one of the most commonly counterfeited. Furthermore, considering watches of a single brand that were seized over a relatively short period of seven months, increased the probability of finding links between them. This was because it is more likely that the same raw materials were used for production of the watches and also that the same tools were used for their tagging. Notably, we wanted to investigate the possibility of using a restricted specimen set to extract larger

market insights through chemical profiling of the watchcases and associating their physical profiling results with links to previously profiled counterfeit watches.

X-ray spectroscopy (XRF) and inductively coupled plasma mass spectrometry (ICP-MS) were the analytical methods used to analyse all the watchcases. Despite the highly satisfactory analytical results obtained, the laborious sample preparation for ICP-MS analysis constitutes a major drawback. Nonetheless, the analytical data allowed us to create reliable and informative profiles; they were subsequently compared and grouped by means of similarity indexes, dividing the specimen set into 15 distinct chemical classes. Several links between specimens were revealed through this process. Even though this method is still in the developmental stage, the proposed approach displays promising capabilities and our results support the chemical profiling of counterfeit watches on a larger scale. Future studies would benefit from an enlarged specimen set; imperatively, the set should contain watches known to originate from common and different production sources. This would allow for the evaluation of the intravariability by computing the distribution of sample pairs from the same production site and intervariability by computing the distribution of sample pairs from unrelated production sites. This in turn would allow the utility and performance of the binary classification model to be either validated or refuted.

While the potential harmful effects of some counterfeit goods are well documented, for watches, there are no published studies regarding the presence of harmful compounds to date. To fill this gap, a second specimen set consisting of 35 replica watches of various brands, that included high-, average- and low-quality counterfeits was also tested using pXRF and a colorimetric test for nickel liberation. In particular, the results showed that the metallic components of the low-end replicas possessed unsafe levels of lead, and the highest lead concentrations were measured in the imitation gemstones; these values exceeded the legal limits as governed by Swiss and European legislation. It was determined that counterfeiters do not comply with safety standards in the alloys that they use. Furthermore, the nickel release test indicated positive results in several parts of the counterfeit watches from the low-quality set. However, these measurements provide little information about actual nickel release and whether it is below or above the legal limit. Although these results are alarming for this type of low-quality

counterfeits, further testing that includes many more specimens is highly advised for a more accurate risk assessment.

In the second part of this work, links between the different watch components that were identified by codification of the physical characteristics of all present imprints were comprehensively studied. The fairly complex interpretation of the relationships between the different watch components may impede a clear understanding of the tagging process. Nevertheless, strong connections between the 35 watches at the physical level were revealed, which indicated that these watches emerged from an interconnected counterfeiting organisation.

While nearly two third of all imprints identified did not generate any links between the 35 watches, they may still be connected to other counterfeit watches previously examined. Hence, all the imprints were then analysed in a more extended context. A retrospective analysis of the FH database was carried out using the 35 watches of the specimen set as a starting point, which resulted in links being found with over 500 other watches. The imprints that generated the most links were then further analysed, it was possible to identify four groups of independent production lines for the straps, as well as groups for the clasps and the crowns. The latter two components generated a great number of links between watches that were not otherwise related based on their imprints. This indicated that they were likely to be produced independently from the other components. In order to further aid the interpretation, a specificity criterion was assigned to each of the imprints, since brand-specific imprints only reveal links between watches of the same brand, and model-specific imprints only link between watches of the same model. The latter was generally found on the dials, but only a few isolated cases revealed a large number of links to previously analysed imprints. In addition, non-specific imprints are also of great interest, since they could also reveal links between counterfeits from different brands. It was found that the brand-specific and non-specific imprints generated most of the links and, thus, are useful to gather information related to possible mass production.

From a temporal perspective, it should be pointed out that several years after the first observation of certain imprints, new replica watches containing imprints produced with the same tagging tools reappeared on the market. Since all watches in the initial specimen set were seized at the Swiss border, they only represent a small portion of the

total number of replica watches seized worldwide and are a minute portion of the entire market. Furthermore, only a selection of the total number of seized watches are examined by the FH. Hence, the imprints that were identified to generate most of the links support the existence of prolific, large-scale production networks. These findings allowed us to design recommendations to optimize the information content that should be gathered during the systematic analysis of imprints.

Overall there was a low correspondence between the chemical and physical links. The chemical profiling of watchcases added knowledge that was previously unknown. Moreover, these links provided information on the production step, which is prior to the tagging process of the different components. Watchcases could thus be produced at several production sites, as supported by the chemical classes, followed by distribution towards multiple, independent assembly sites. In some cases, the chemical links were confirmed by the physical ones. This is mainly true for watches with a high visual similarity (same brand, model and plating), which were confiscated at the same time and place. It is thus likely that they were purchased at the same time and originated from the same production and assembly source. While this may be true, chemically linked watches without physical links, and physically linked watches without chemical links were also found. This in turn again supports the hypothesis of a large-scale organized network.

In summary, it can be emphasised that each source of information (chemical, physical, temporal and geographical) investigated during this work resulted in new insights on the counterfeiting process. Despite the limited number of specimens initially considered, we were able to reveal or confirm links between watches that were previously unknown or uncertain; demonstrate the interconnection of all watches on a chemical and/or physical level; and finally, through analysis of the data, suggest the presence of an overarching organised network with substructures. The proposed approach, oriented from a forensic intelligence perspective, allowed us to combine the different dimensions in an interdisciplinary manner. The resulting insights provide valuable investigative leads and a solid basis for future research. Depending on the level of intelligence, focus could be placed on understanding and preventing watch counterfeiting, investigating leads for a particular case, or, more broadly, for establishing the presence of an organized trafficking network.

Conclusion

Our research exemplifies a multi-disciplinary approach which demonstrates how forensic science can actively contribute to the gathering of intelligence on illicit markets.

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11 Annex

11.1 Specimen set 1

| | | |
|---|--|-----------------------------|
| ID: M01 (15916) | Model: Deepsea (Sea-Dweller) | |
|  |  | |
| Period/ Date of seizure: 01.09.2015 | | |
| Fabrication: China | Provenance: Thailand | Seizure: Switzerland |
| | Destination: Italy | |

| | | |
|---|--|-----------------------------|
| ID: M02 (16008-1) | Model: Submariner Date | |
|  |  | |
| Period/ Date of seizure: March-April 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | |

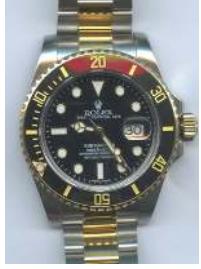
| | | |
|---|--|-----------------------------|
| ID: M03 (16008-2) | Model: Cosmograph Daytona | |
|  |  | |
| Period/ Date of seizure: March-April 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | |

| | | |
|---|--|--------------------------|
| ID: M04 (16018-1) | Model: Cosmograph Daytona | |
|  1 |  2 | |
| Period/ Date of seizure: 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Chiasso, |
| | Destination: - | Switzerland |

| | | |
|--|---|--------------------------|
| ID: M05 (16018-2) | Model: Submariner Date | |
|  2 |  2 | |
| Period/ Date of seizure: 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Chiasso, |
| | Destination: - | Switzerland |

| | | |
|---|--|---------------------------------|
| ID: M06 (16029-1) | Model: Submariner Date | |
|  1 |  1 | |
| Period/ Date of seizure: 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Zurich Airport, |
| | Destination: - | Switzerland |

| | | |
|---|--|---------------------------------|
| ID: M07 (16029-2) | Model: Submariner Date | |
|  |  | |
| Period/ Date of seizure: 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Zurich Airport, |
| | Destination: - | Switzerland |

| | | |
|--|---|---------------------------------|
| ID: M08 (16030) | Model: Submariner Date | |
|  |  | |
| Period/ Date of seizure: 2015 | | |
| Fabrication: China | Provenance: - | Seizure: Zurich Airport, |
| | Destination: - | Switzerland |

| | | |
|---|--|------------------------------|
| ID: M09 (16093-1) | Model: Cosmograph Daytona | |
|  |  | |
| Period/ Date of seizure: October 2015 | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, |
| | China/Singapore/China | Switzerland |

| | |
|---|--|
| ID: M10 (16093-2) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | |
|--|---|
| ID: M11 (16094-1) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | |
|---|--|
| ID: M12 (16094-2) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | |
|---|--|
| ID: M13 (16095-2) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | |
|--|---|
| ID: M14 (16096-1) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | |
|---|--|
| ID: M15 (16096-2) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | | |
|---|--|--|
| ID: M16 (16102-1) | Model: Datejust | |
|  |  | |
| Period/ Date of seizure: October 2015 | | |
| Fabrication: China | Provenance: China/Singapore/China | Seizure: Zurich Post, Switzerland |
| | | |

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|--|---|--|
| ID: M17 (16102-2) | Model: Datejust | |
|  |  | |
| Period/ Date of seizure: October 2015 | | |
| Fabrication: China | Provenance: China/Singapore/China | Seizure: Zurich Post, Switzerland |
| | | |

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|---|--|--|
| ID: M18 (16103-1) | Model: Submariner Date | |
|  |  | |
| Period/ Date of seizure: October 2015 | | |
| Fabrication: China | Provenance: China/Singapore/China | Seizure: Zurich Post, Switzerland |
| | | |

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|---|--|---|-------------|
| ID: M19 (16103-2) | Model: Submariner Date | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

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|--|---|---|-------------|
| ID: M20 (16105-2) | Model: Datejust | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

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|---|--|---|-------------|
| ID: M21 (16106-2) | Model: Daytona | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

| | | | |
|---|--|---|-------------|
| ID: M22 (16107) | Model: Air-King | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

| | | | |
|--|---|---|-------------|
| ID: M23 (16104-1) | Model: Day-Date | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

| | | | |
|---|--|---|-------------|
| ID: M24 (16104-2) | Model: Day-Date | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

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|---|--|---|-------------|
| ID: M25 (16095-1) | Model: Cosmograph Daytona | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

| | | | |
|--|---|---|-------------|
| ID: M26 (16097-1) | Model: Cosmograph Daytona | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

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|---|--|---|-------------|
| ID: M27 (16097-2) | Model: Cosmograph Daytona | | |
|  |  | | |
| Period/ Date of seizure: October 2015 | | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China | Switzerland |

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|---|--|
| ID: M28 (16098-1) | Model: Day-Date |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: |
| | Seizure: Zurich Post, China/Singapore/China |
| | Switzerland |

| | |
|--|---|
| ID: M29 (16098-2) | Model: Day-Date |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: |
| | Seizure: Zurich Post, China/Singapore/China |
| | Switzerland |

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|---|--|
| ID: M30 (16099-1) | Model: Day-Date |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: |
| | Seizure: Zurich Post, China/Singapore/China |
| | Switzerland |

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|---|--|
| ID: M31 (16099-2) | Model: Day-Date |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

| | |
|--|---|
| ID: M32 (16100-1) | Model: Day-Date |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

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|---|--|
| ID: M33 (16100-2) | Model: Cosmograph Daytona |
|  |  |
| Period/ Date of seizure: October 2015 | |
| Fabrication: China | Provenance: China/Singapore/China |
| | Seizure: Zurich Post, Switzerland |

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|---|--|---|
| ID: M34 (16105-1) | Model: Datejust | |
|  |  | |
| Period/ Date of seizure: October 2015 | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China |
| | | Switzerland |

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|--|---|---|
| ID: M35 (16106-1) | Model: Yacht-Master II | |
|  |  | |
| Period/ Date of seizure: October 2015 | | |
| Fabrication: China | Provenance: | Seizure: Zurich Post, China/Singapore/China |
| | | Switzerland |

11.2 Specimen set 2

| | | |
|---|--|------------------------------|
| ID: M01 (15405) | Brand: Hublot | Model: Classic Fusion |
|  |  | |
| Period/ Date of seizure: 17.03.2014 | | Quality: High |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|--|---|-----------------------------|
| ID: M02 (15412-1) | Brand: Hublot | Model: Big Bang |
|  |  | |
| Period/ Date of seizure: 17.03.2014 | | Quality: High |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|---|--|------------------------------|
| ID: M03 (15404-2) | Brand: Hublot | Model: Classic Fusion |
|  |  | |
| Period/ Date of seizure: 17.03.2014 | | Quality: High |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|---|--|-----------------|
| ID: M04 (15417-1) | Brand: Patek Philippe | Model: - |
|  |  | |

| | |
|--|-----------------------------|
| Period/ Date of seizure: 17.03.2014 | Quality: High |
| Fabrication: China | Provenance: - |
| | Seizure: Switzerland |
| | Destination: - |
| | Police seizure |

| | | |
|--|---|-----------------|
| ID: M05 (15417-2) | Brand: Patek Philippe | Model: - |
|  |  | |

| | |
|--|-----------------------------|
| Period/ Date of seizure: 17.03.2014 | Quality: High |
| Fabrication: China | Provenance: - |
| | Seizure: Switzerland |
| | Destination: - |
| | Police seizure |

| | | |
|---|--|-----------------------|
| ID: M06 (15433-2) | Brand: Tag Heuer | Model: Carrera |
|  |  | |

| | |
|--|-----------------------------|
| Period/ Date of seizure: 17.03.2014 | Quality: High |
| Fabrication: China | Provenance: - |
| | Seizure: Switzerland |
| | Destination: - |
| | Police seizure |

| | | |
|---|--|-----------------------------|
| ID: M07 (15448-1) | Brand: Audemars Piguet | Model: Royal Oak |
|  |  | |
| Period/ Date of seizure: 17.03.2014 | | Quality: High |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|--|---|-----------------------------|
| ID: M08 (15448-2) | Brand: Audemars Piguet | Model: Royal Oak |
|  |  | |
| Period/ Date of seizure: 17.03.2014 | | Quality: High |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|---|--|-----------------------------|
| ID: M09 (15432-1) | Brand: Tag Heuer | Model: Diamond Star |
|  |  | |
| Period/ Date of seizure: 17.03.2014 | | Quality: High |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|---|-------------------------|--|
| ID: M10 (15432-2) | Brand: Tag Heuer | Model: Diamond Star |
|  | |  |
| Period/ Date of seizure: 17.03.2014 | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Police seizure |

| | | |
|--|-------------------------|---|
| ID: M11 (14978) | Brand: Breitling | Model: Bentley |
|  | |  |
| Period/ Date of seizure: - | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: Switzerland |
| | Destination: - | Geneva Airport |

| | | |
|---|-------------------------|--|
| ID: M12 (14424) | Brand: Breitling | Model: Bentley |
|  | |  |
| Period/ Date of seizure: 20.06.2011 | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: Greece, |
| | Destination: - | Athens, Police Seizure |

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|---|--|------------------------------|
| ID: M13 (FH inventory) | Brand: Hublot | Model: Classic Fusion |
|  |  | |

| | |
|-----------------------------------|-------------------------|
| Period/ Date of seizure: - | Quality: Average |
| Fabrication: - | Provenance: - |
| | Seizure: - |
| | Destination: - |

| | | |
|--|---|------------------------------|
| ID: M14 (14586-2) | Brand: Hublot | Model: Classic fusion |
|  |  | |

| | |
|--|---------------------------|
| Period/ Date of seizure: 05.10.2012 | Quality: Average |
| Fabrication: China | Provenance: - |
| | Seizure: Paraguay, |
| | Destination: - |
| | Asuncion, Police seizure |

| | | |
|---|--|--------------------------|
| ID: M15 (14587) | Brand: Hublot | Model: King Power |
|  |  | |

| | |
|--|---------------------------|
| Period/ Date of seizure: 05.10.2012 | Quality: Average |
| Fabrication: China | Provenance: - |
| | Seizure: Paraguay, |
| | Destination: - |
| | Asuncion, Police Seizure |

| | | |
|---|-----------------------|------------------------------|
| ID: M16 (FH inventory) | Brand: Hublot | Model: Classic Fusion |
|  | | |
| Period/ Date of seizure: - | | Quality: Average |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|--|-----------------------------|---------------------------|
| ID: M17 (14604) | Brand: Ulysse Nardin | Model: Marin |
|   | | |
| Period/ Date of seizure: 05.10.2012 | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: Paraguay, |
| | Destination: - | Asuncion, Police seizure |

| | | |
|--|-----------------------------|-------------------------|
| ID: M18 (FH inventory) | Brand: Ulysse Nardin | Model: Marin |
|   | | |
| Period/ Date of seizure: - | | Quality: Average |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|---|------------------------------|--|
| ID: M19 (14585) | Brand: Patek Philippe | Model: - |
|  | |  |
| Period/ Date of seizure: 05.10.2012 | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: Paraguay, Asuncion, Police seizure |
| | Destination: - | |

| | | |
|--|-----------------------|---|
| ID: M20 (14775) | Brand: Rolex | Model: Datejust |
|  | |  |
| Period/ Date of seizure: October 2012 | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: United Arab Emirates, Dubai, Police investigation |
| | Destination: - | |

| | | |
|---|-------------------------|--|
| ID: M21 (14600) | Brand: Tag Heuer | Model: Mikrotourbillon |
|  | |  |
| Period/ Date of seizure: 05.10.2012 | | Quality: Average |
| Fabrication: China | Provenance: - | Seizure: Paraguay, Asuncion, Police seizure |
| | Destination: - | |

| ID: M22 (14369) | Brand: Frank Müller | Model: Elegance |
|---|--|-----------------|
|  |  | |

Period/ Date of seizure: 05.04.2012

Quality: Average

Fabrication: China

Provenance: -

Seizure: United Arab Emirates, Dubai, Police seizure

Destination: -

| ID: M23 (14980) | Brand: Frank Müller | Model: Crazy Color Dreams |
|--|---|---------------------------|
|  |  | |

Period/ Date of seizure: -

Quality: Average

Fabrication: China

Provenance: -

Seizure: Switzerland, Geneva Airport

Destination: -

| ID: M24 (15056-1) | Brand: Tissot | Model: PRC 200 |
|---|--|----------------|
|  |  | |

Period/ Date of seizure: 31.12.2010

Quality: Low

Fabrication: China

Provenance: -

Seizure: United Arab Emirates, Dubai

Destination: -

| ID: M25 (15059-2) | Brand: Tag Heuer | Model: - |
|-------------------------------------|------------------|--------------------------------------|
| | | |
| Period/ Date of seizure: 31.12.2010 | Quality: Low | |
| Fabrication: China | Provenance: - | Seizure: United Arab Emirates, Dubai |
| | Destination: - | |

| ID: M26 (15072-2) | Brand: Chopard | Model: - |
|-------------------------------------|----------------|--------------------------------------|
| | | |
| Period/ Date of seizure: 31.12.2010 | Quality: Low | |
| Fabrication: China | Provenance: - | Seizure: United Arab Emirates, Dubai |
| | Destination: - | |

| ID: M27 (15063-2) | Brand: Hermès | Model: Arceau |
|-------------------------------------|----------------|--------------------------------------|
| | | |
| Period/ Date of seizure: 31.12.2010 | Quality: Low | |
| Fabrication: China | Provenance: - | Seizure: United Arab Emirates, Dubai |
| | Destination: - | |

| | | |
|---|------------------------|--|
| ID: M28 (9502) | Brand: Longines | Model: Grande Classique |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|--|-------------------------------|---|
| ID: M29 (FH inventory) | Brand: Audemars Piguet | Model: - |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|---|-----------------------|--|
| ID: M30 (6760) | Brand: Omega | Model: Constellation |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|---|-----------------------|--|
| ID: M31 (FH inventory) | Brand: Rolex | Model: Datejust |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|--|-----------------------|---|
| ID: M32 (4984) | Brand: Rolex | Model: Submariner |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|---|------------------------------|--|
| ID: M33 (15073-2) | Brand: Patek Philippe | Model: Sky Moon |
|  | |  |
| Period/ Date of seizure: 31.12.2010 | | Quality: Low |
| Fabrication: China | Provenance: - | Seizure: United Arab Emirates, Dubai |
| | Destination: - | |

| | | |
|---|-----------------------|--|
| ID: M34 (FH inventory) | Brand: Gucci | Model: - |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

| | | |
|--|-----------------------|---|
| ID: M35 (6858) | Brand: Gucci | Model: - |
|  | |  |
| Period/ Date of seizure: - | | Quality: Low |
| Fabrication: - | Provenance: - | Seizure: - |
| | Destination: - | |

ISBN 2-940098-89-1