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RESEARCH ARTICLE

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European ceramic technology in the Far East: enamels and pigments in Japanese art from the 16th to the 20th century and their reverse influence on China

Riccardo Montanari^{1*} , Nobuyuki Murakami², Philippe Colomban³, Maria Francesca Alberghina⁴, Claudia Pelosi⁵ and Salvatore Schiavone⁴

Abstract

The production of Japanese enamels for porcelain decoration was thought to have originated from the direct and exclusive influence of Chinese potters who moved to Japan during the chaotic Ming to Qing dynastic change in 1644. Recent systematic studies have identified, for the first time, the crucial influence of Jesuit missionaries on pigment and enamel production in Japan from the late 16th-century. In particular, such first encounter laid the foundation for the continued influence exerted by European technology on Japanese art throughout the centuries. The present study has further identified European enamels used for the decoration of polychrome wares fired in Arita, the porcelain production center of Japan. This continued exchange not only marked the Edo period, but also extended into the twentieth century. For the first time, the lack of written records regarding the use of western pigments for enamel production caused by the persecutions of European and Japanese Christians has been overcome in the work herein presented. The nature of the imported materials has been firmly identified and characterized. The analytical results (EDXRF and Raman) have finally revealed how western technology and materials not only kept influencing Japanese art during the isolation (sakoku) period, but also accompanied the strong westernization process that marked Japanese history from the late nineteenth century. Moreover, the significant reverse influence of Japanese-made enamels on Chinese polychrome porcelain production in the late Qing and twentieth century has been fully identified for the first time. Furthermore, results show that the shift of the Pb mode of lead antimonate (Naples Yellow) is affected by the firing temperature for enamel decoration, and that this characteristic, along with the chemical composition, enables the identification of the origin and manufacture period of the yellow enamel.

Keywords: Porcelain, Enamel, Pigment, Painting, Japan, Jesuit, Seminario, China, EDXRF, Raman

Introduction and historical context

The production of overglaze enamels in Japan was long thought to have originated from the direct and exclusive influence of Chinese potters in the mid seventeenth century [1, 2] when the collapse of the Ming dynasty

caused many of them to flee to Japan to find new profitable markets for their own survival. Yet, recent systematic studies on the origin of the materials employed for polychrome decoration identified, for the first time, the technological exchange that occurred between Jesuit missionaries and Japanese painters and potters from the late sixteenth century [3, 4]. In particular, the European Renaissance practice of using the same coloring agents both for paintings and ceramics was unquestionably confirmed by the matching compound detected on a

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coloured porcelain and a European style painting created by a Japanese painter who studied at the Jesuit *Seminario* under the direction of the Italian Jesuit painter Giovanni Cola from 1583 [4]. The production of sacred and secular images based on models imported from Europe, be it prints, engravings or paintings, involved the use of European pigments that did not exist in Japan at the time [3–6]. These new pigments also marked Chinese enamel production from the early eighteenth century, after Jesuit missionaries were ordered by the Kangxi Emperor in 1695 to establish a glass workshop in the Palace to reproduce the enamels of western origin that had strongly impressed him [7, 8]. Recently, Raman analyses have confirmed the use of European enamel precursors at the end of Kangxi reign as mentioned in historical records [9, 10]. This pattern of influence had actually originated in Japan in the late sixteenth century [3, 4]. Additional evidence comes from *Naito Tokuen Johan* [11], a Christian samurai and close friend of Takayama Ukon, who supported the establishment of a glass workshop in the painting Seminario built by the Jesuits in Arita in 1595 under the supervision of Giovanni Cola [11]. The Jesuit Seminario was attended by Japanese and Chinese painters eager to learn European oil painting techniques [12]. The imported compounds employed for such purpose were scarce and very expensive [6], so their use was strictly allowed to those having trained under the direct supervision of Giovanni Cola himself. In particular, tin and antimony were important elements used in Renaissance Europe for enameling of majolica [13–16], Limoges metal wares [17], and for preparing painting pigments [18, 19]. Both end-members of pyrochlore solid solutions (lead–tin yellow and lead antimonate yellow) as well as more complex solid solutions had been largely used since Roman times [20–22].

These same pigments have been detected on the famous *Painting of the Madonna with the Infant Jesus and Her Fifteen Mysteries* (1620 s, Japan) [5] and on the rare *mukozuke* dish bearing the Japanese mark '*Kan'ei Nen Sei*' (made in the *Kan'ei* period, that is 1624–1644) [3]. Those instances have provided the definitive proof that the first phase of technological exchange between Europe and Japan started in the late sixteenth century, and that it continued until the final expulsion of all Christians by the Tokugawa Shogunate in 1639 as a result of the strictly enforced persecutions, with no surviving documents relating to the Jesuit presence and the early interaction with the Japanese. This study has proven, for the first time, how such first technological exchange between Jesuit missionaries and the Japanese in the sixteenth century laid the foundation for the continued influence exerted by Europeans on Japanese applied arts (porcelain,

prints, and paintings) throughout the Edo period (1603–1868) and into the twentieth century.

We report here a non-destructive analysis carried out by X-Ray Fluorescence (ED-XRF) and Raman spectroscopy in order to get more information on the use of these ceramic pigments, thus overcoming the daunting issue of the lack of written records regarding the use of western pigments and/or western technology for enamel production. Shards of polychrome decorated porcelains fired in Arita, the porcelain production center of Japan, dating from the nineteenth and twentieth century, were analyzed with the aim of identifying the coloring agents employed for such production. The results have finally provided a definitive answer, revealing how western technology and materials not only kept influencing Japanese art during the isolation (*sakoku*) period, but also accompanied the strong westernization process of the country that marked Japanese craft and industry from the late nineteenth century.

Materials and methods

Analyzed shards

All analyzed shards were provided by the Arita Museum of History, Arita, Saga Prefecture, Japan, and were mostly excavated at the *Aka-e machi* site (enamellers' quarter) located in the *Uchiyama* area of Arita, where all enamellers had been moved by the Nabeshima clan in the late 1650s in order to improve the production process through better labor organization [4]. The shards provide crucial information regarding the coloring agents and enamel preparation techniques employed when the presence of western countries increased significantly and the Tokugawa shogunate was about to collapse. Characteristics are summarized in Fig. 1.

XRF spectrometer: experimental and measurement parameters

The employed XRF portable instrument consists of a miniature X-ray tube system (Amptek, X-123 SDD, Mini-X X-ray tube, USA) which includes the X-ray tube (max voltage of 40 kV, max current of 0.2 mA, target Rh, collimator 1 or 2 mm), the power supply, the control electronics and the USB communication for remote control; a Silicon Drift Detector (SDD) with a 125 to 140 eV FWHM @ 5.9 keV Mn K α line Energy Resolution (depends on peaking time and temperature); 1 keV to 40 keV detection range of energy; max rate of counts to 5.6×10^5 cps; software for acquiring and processing the XRF spectra.

Primary beam and detector axis form an angle of 0 and 40 degrees respectively with the perpendicular to the sample surface. Measurement parameters were as follows: tube voltage 35 kV; current 80 μ A, acquisition time 100 s; no filter was applied between the X-Ray tube and the sample;

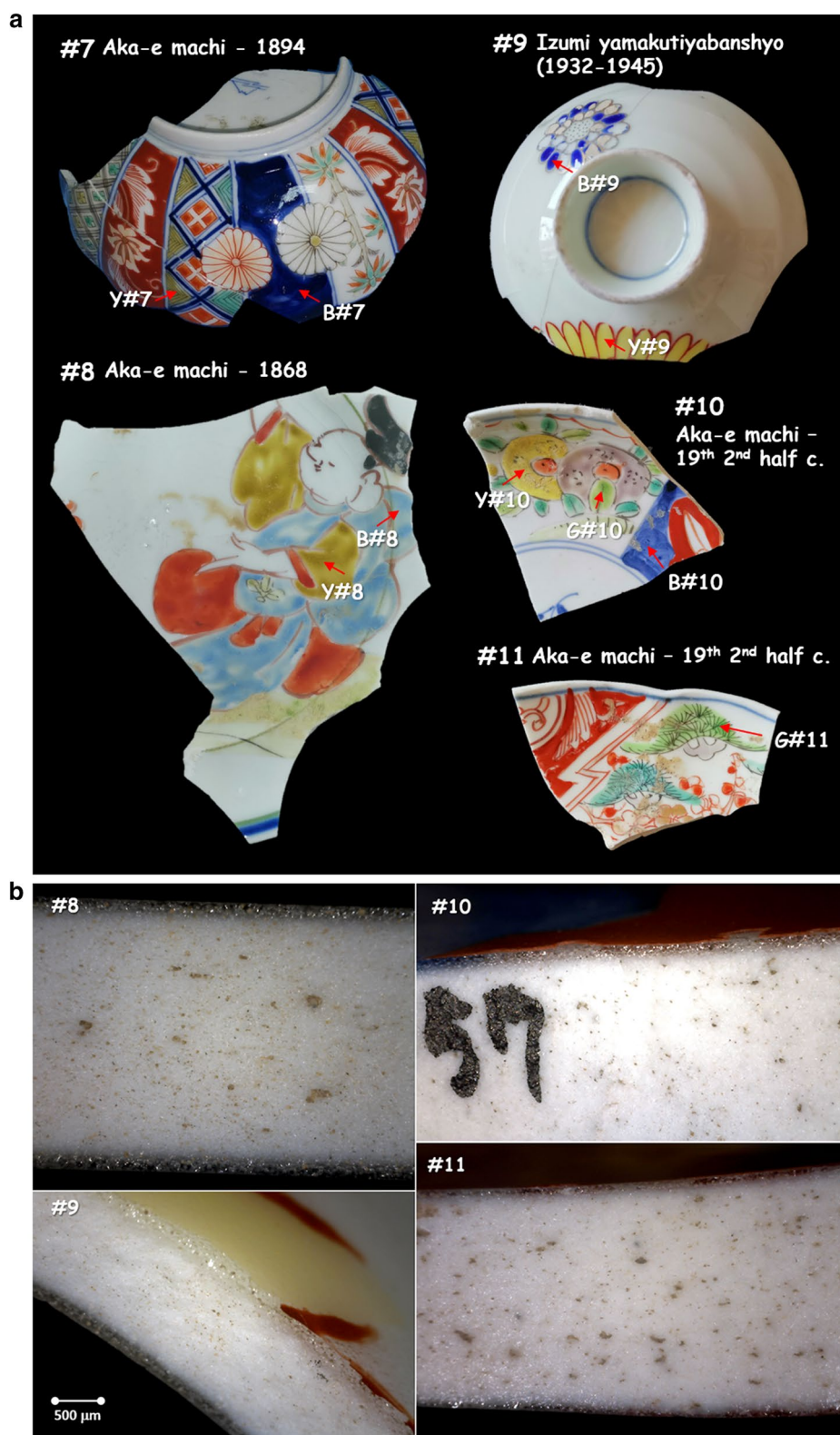


Fig. 1 **a** Analyzed shards with underglaze and overglaze decoration. Kiln sites, production date and XRF or Raman measurement areas are given (the dimensions are here reported: Shard #7: 12 × 9 cm²; #8: 16 × 13 cm²; #9 diameter: ~9.8 cm; #10: 9.5 × 8 cm²; #11: 7 × 5 cm²). **b** Fracture sections showing the glaze-body interface of the #8, #9, #10, #11 analysed shards acquired by using portable optical microscopy (50 magnification) along the fracture lines without any sampling

distance between sample and detector around 1 cm. The setup parameters were selected to have a good spectral signal and to optimize the signal to noise ratio (SNR). Measurement data were processed using the factory provided data reduction DPPMCA software, which enables the acquisition, display, and control for Amptek signal processors and allows the automatic peak recognition supported by manual peak selection and checking. The software further enables curve fitting based on chosen elements to ensure a match between the measured spectra and theoretically predicted spectra calculated from fundamental parameters (FP). Finally, XRF spectra have been graphically provided by Origin Pro 8.5.

Raman spectroscopy

The micro-Raman analysis was performed through a Renishaw InVia Raman Microscope (UK). The fragments were examined without any preparation. In order to make the fragment surface perfectly perpendicular to the laser path and to improve the focus, the shards were positioned on a clay base. Two exiting wavelengths were used: 532 nm and 633 nm. The diffused light was recorded in a backscattered geometry using a 50× objective at long focal distance, in order to obtain a measurement spot of about 4 microns in diameter. Spectra were managed by Wire 2.0 software, associated to the Renishaw instrument.

Results and discussion

The overglaze enameled shards analyzed in this study are characterized by the presence of coloring agents of European origin. We will discuss the Raman and XRF data recorded on the different colored areas, yellow, blue, and green.

Overglaze yellow enamels

Raman analysis

The Raman spectra detected on the nineteenth and twentieth century Japanese yellow enamels and on the seventeenth century *mukozuke* dish are reported in Fig. 2.

Naples yellow pyrochlore is a non-stoichiometric $A_{2-x}A'B_{2-y}B'_yO_{7-\delta}$ phase built with two sub-lattices, one of (big) A cations ($A = Pb^{2+}$ in the case of Naples yellow pigment), the second of (small, covalent bonded) cations ($B = Sb, Sn, Fe, Zn, Si, Zr...$) forming tetrahedral and octahedral entities sharing some oxygen atoms [23, 24]. B cations are easily oxidized/reduced and thus the formula and oxygen stoichiometry depend on the firing atmosphere. Due to the high mass of lead, the Raman signature show a characteristic strong peak arising from the Pb translational mode at low wavenumber, ranging from ~ 125 to $\sim 140\text{ cm}^{-1}$ as a function of the composition/structure [13–15, 17, 25]. Studies of Sn–O and Sb–O end members have allowed the identification of the Sn–O

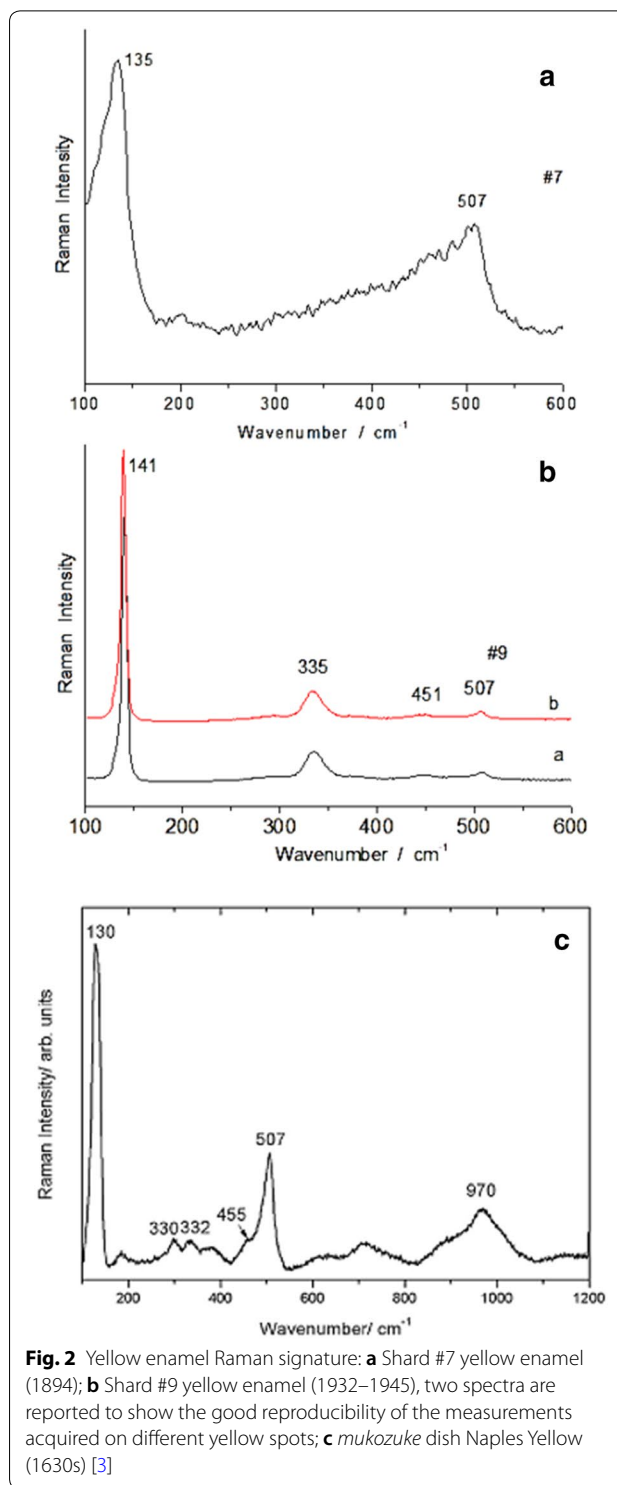


Fig. 2 Yellow enamel Raman signature: **a** Shard #7 yellow enamel (1894); **b** Shard #9 yellow enamel (1932–1945), two spectra are reported to show the good reproducibility of the measurements acquired on different yellow spots; **c** *mukozuke* dish Naples Yellow (1630s) [3]

and Sb–O stretching modes (stretching modes are strong in Raman spectroscopy) at ca. 450 and 505 cm^{-1} respectively [17]. Thus differentiation between Sn-rich and Sb-rich Naples Yellow is obvious from their Raman spectrum: Sb-rich pigment shows two strong peaks at ca. 130

and 505 cm^{-1} , while Sn-rich pigment shows a spectrum with peaks at $\sim 130, 325, 450$ and 505 cm^{-1} [17].

As shown in Fig. 2, the spectra recorded on the yellow decorations of shards #7, #9, and the previously analysed seventeenth century *mukozuke* porcelain, all show a strong ca. $130\text{--}140\text{--}505\text{ cm}^{-1}$ doublet, characteristic of a Sb-rich Naples Yellow pyrochlore. However, the three yellow enamels feature different wavenumber positions of the strong Pb mode, namely $135, 141$ and 130 cm^{-1} , and all prove to be a shift higher than the uncertainty of the measure.

The lowest value detected on the *mukozuke* dish, 130 cm^{-1} , corresponds to a firing temperature in the range of $900\text{--}1000\text{ }^\circ\text{C}$ according to Sakellariou et al. [16]. Such temperature value is consistent with the firing temperatures that characterize lead antimonate yellows in Renaissance Italy [13, 16, 18, 26–30].

XRF analysis

XRF spectra recorded on yellow colored areas show strong Pb peaks and medium Si peaks as expected for lead-based overglaze enamels.

The Pb-Sb yellow detected on shard #7 (Fig. 3a) and the Pb-Fe-Sb-(Zn) yellow detected on shard #9 (Fig. 3b) both bear an enamel matrix consisting of the ternary system Pb-Si-K. Such matrix composition, along with the presence of antimony, strongly points to the recipe reported by Takamatsu in 1878 [31]: *Shiratama* (PbO, SiO₂, K₂O) and *Tojirome*, an antimony-based compound, are listed as the main components to be used for yellow decoration “...nearly pure antimony... also called *Toshirome*...is used as a yellow enamel with *Shiratama* and other bodies, whereby oxides of antimony and lead are formed which give the yellow colour”. This description confirms that the preparation of the antimony-based enamel differs from the European Renaissance recipe. The Japanese yellow color is not the specific compound obtained through calcination as the traditional European method employed for Naples Yellow required [16, 18], but it appears to be basically a by-product of the reaction between the mixed reagents. Comparison with the spectrum of the Italian Renaissance *Naples Yellow* (lead-antimonate yellow) detected on the famous *mukozuke* dish dated 1624–1644 (Fig. 3c) [3] clearly shows that the late Renaissance enamel consists instead of a Pb-Si matrix with no significant amount of potassium: the yellow on the *mukozuke* dish perfectly matches the recipe for *Giallo de’ Vasari* (Potters’ yellow) published by Valerio Mariani in 1620 in the treatise *Della Miniatura* [13, 14].

The Pb-Cr yellows detected on shards #8 and #10 (Fig. 4a, b) prove to belong to the Cr-based enamels developed in Europe in the early nineteenth century. The use of Cr-based enamels for ceramic decoration was

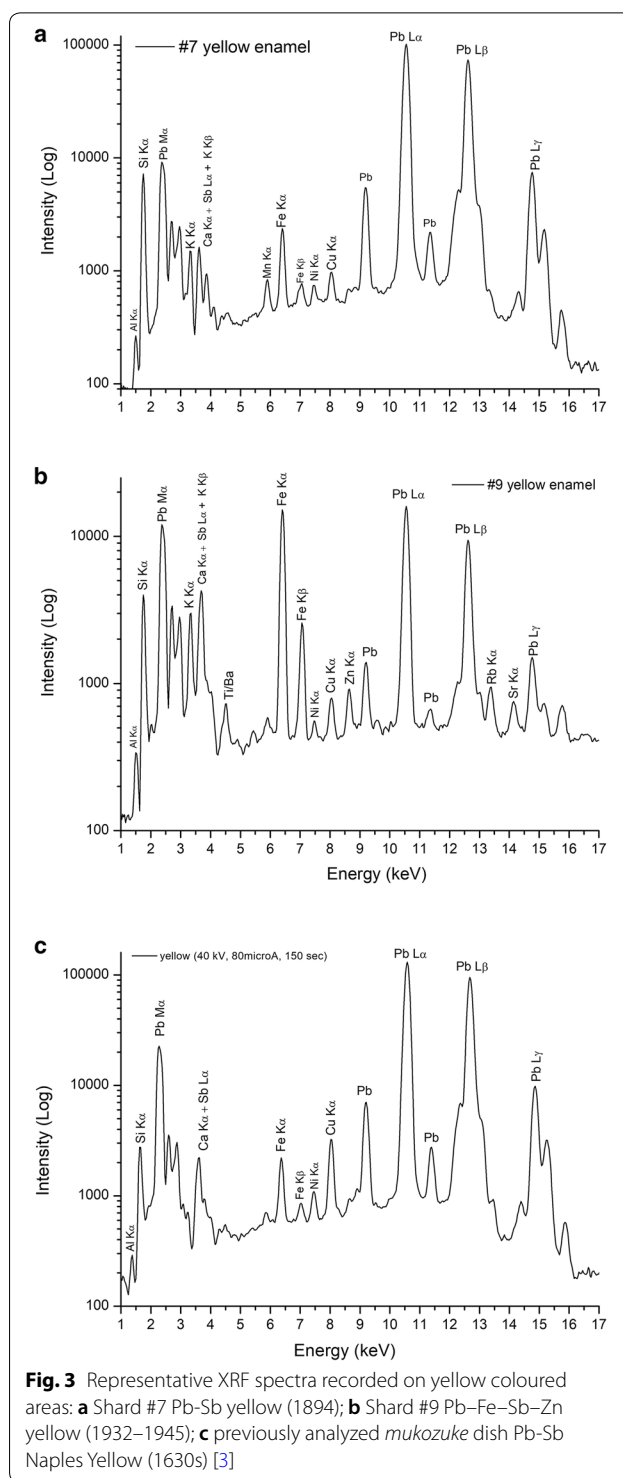
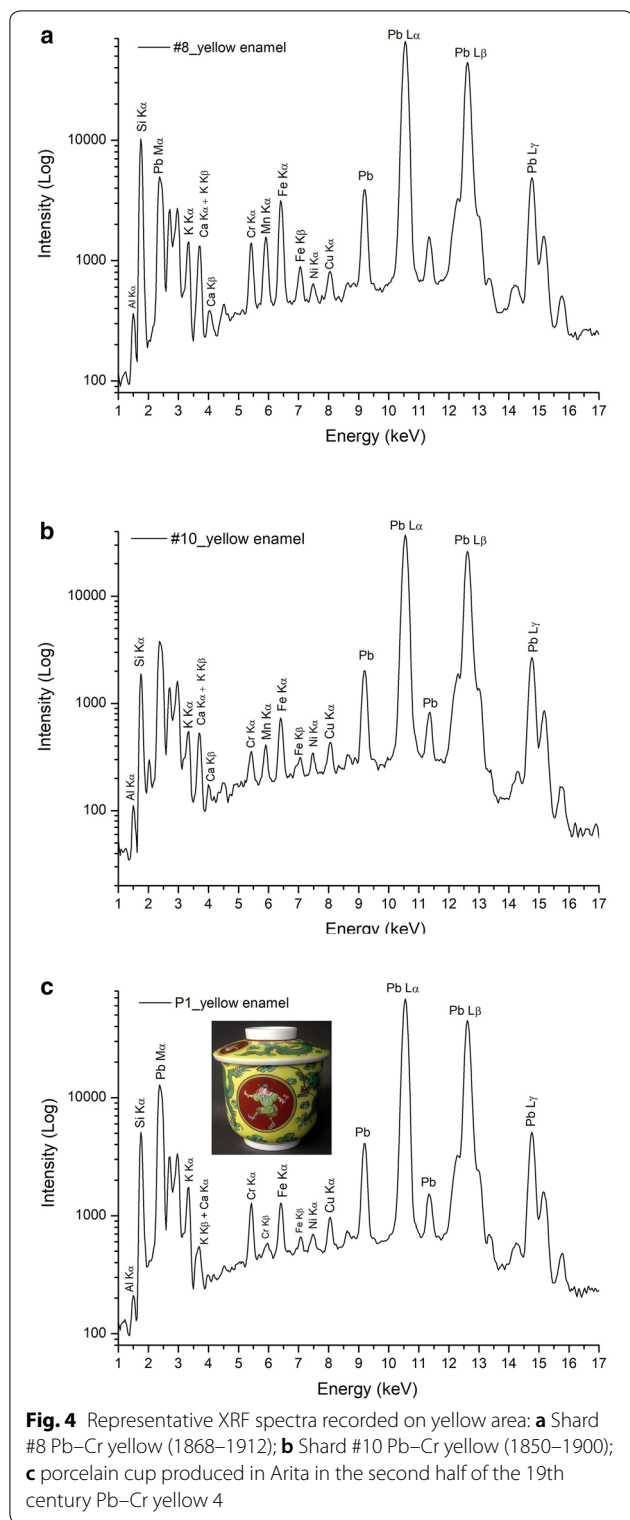


Fig. 3 Representative XRF spectra recorded on yellow coloured areas: **a** Shard #7 Pb-Sb yellow (1894); **b** Shard #9 Pb-Fe-Sb-Zn yellow (1932–1945); **c** previously analyzed *mukozuke* dish Pb-Sb Naples Yellow (1630s) [3]

emphasized by Vauquelin, the discoverer of the element chromium [32]. These new findings are consistent with the previous detection of a Cr-based yellow enamel on a porcelain cup produced in Arita in the second half of the nineteenth century [4] (Fig. 4c), thus confirming the



incorporation of such chromophore in the Japanese porcelain industry in the early-to-mid nineteenth century.

Blue coloring agent and overglaze enamels

The XRF spectrum of the underglaze blue pigment detected on shard #7 shows a manganese to cobalt ratio ($Mn/Co < 1$) incompatible with the use of a natural and pure Asian cobalt ore [33, 34]. This demonstrates that the chromophore consists of a mixture of natural cobalt imported from China and a synthetic blue material developed in Europe (Fig. 5a). It is interesting to note here that the practice of mixing different grades of blue materials in Japan started in the mid-seventeenth-century when trade with Europe became the main drive of the porcelain industry in Arita: western taste demanded heavy decoration and Japanese potters needed to save up on material cost in order to improve profitability [4].

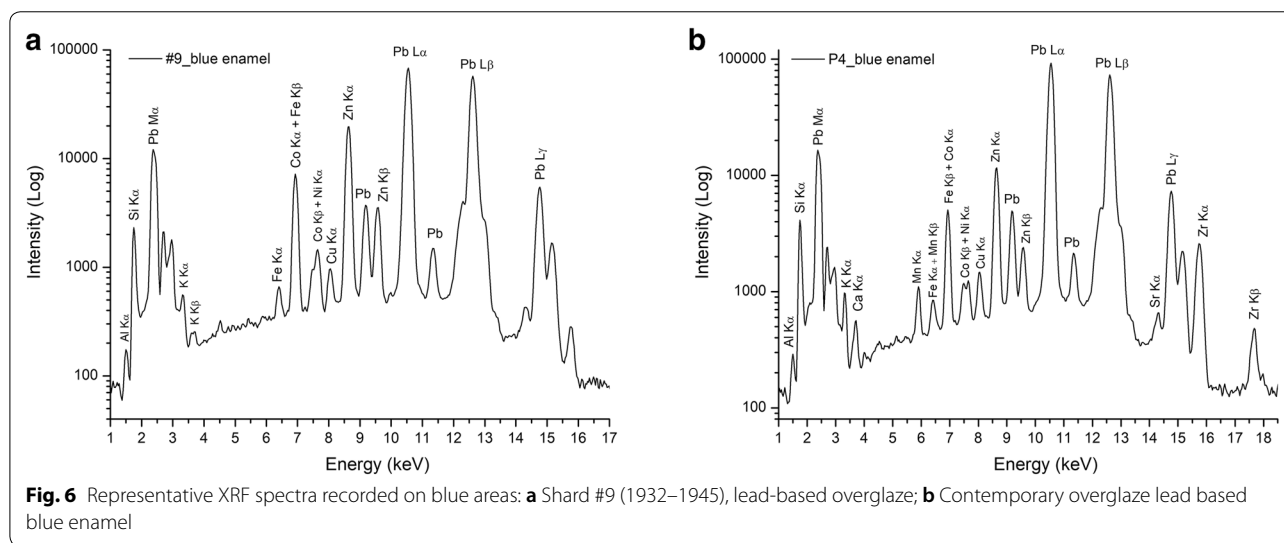
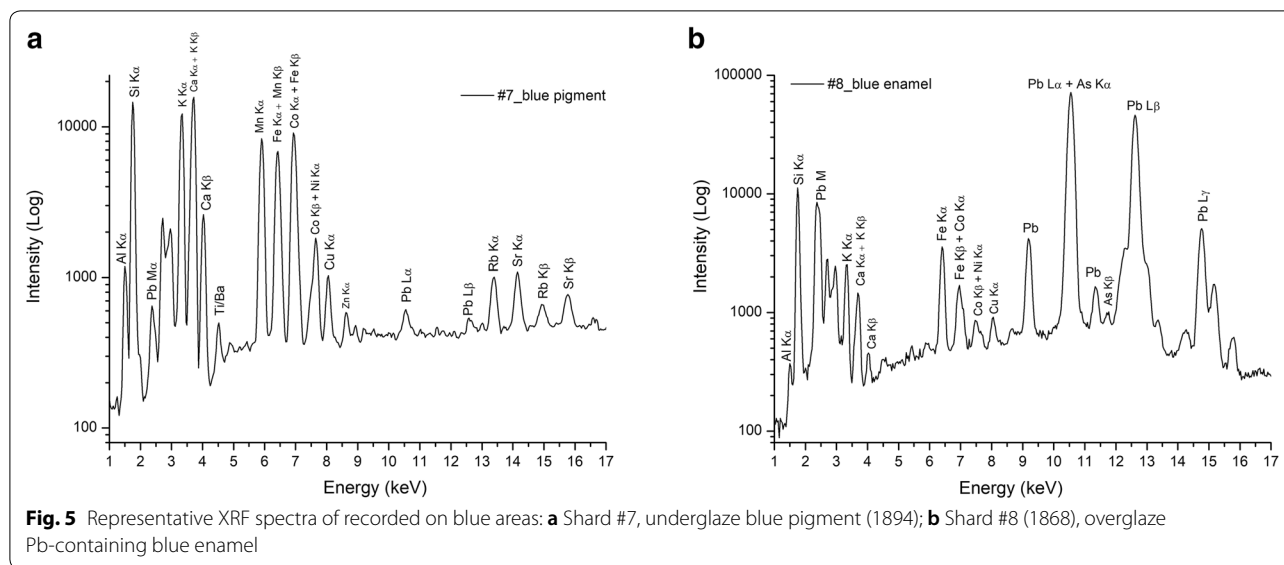
The XRF spectrum recorded on the overglaze blue decoration of shard #8 (Fig. 5b) shows a chemical composition consistent with the European smalt-based enamel characterized by the elements Ni–Fe–As in association with Co. European sources of cobalt are vein of arsenates secondary deposits, while Asian cobalt sources are primary deposits rich in Fe and Mn [35]. The XRF spectrum recorded on shard #9 shows instead the use of an overglaze blue enamel based on a compound of Co and Zn (Fig. 6a).

The results unquestionably prove that starting from the second half of nineteenth century and well into the twentieth century, Japanese potters gradually replaced most of the low-grade natural cobalt ores (rich in manganese and iron) imported from China and the smalt-based overglaze blue enamel imported from the Old Continent with Co-based synthetic pigments of European origin characterized by a higher coloring power and less sensitivity to firing atmosphere. This same type of blue material appears to have been consistently used until the present day as testified by the analysis carried out on a contemporary polychrome porcelain cup of high artistic value fired in Arita (Fig. 6b).

Overglaze green enamels

Visual inspection of the green colored shards herein analyzed strongly points to the intentional use of different compounds aimed at achieving various chromatic effects. Besides the expected use of copper ions dissolved in the glassy matrix for the traditional green color [7], a Cu–Cr-containing enamel was detected on shard #10 by XRF (Fig. 7a). Such instance is in perfect agreement with the recipe published in 1869 by Thomas Salter in *Field's Chromatography or Treatise on Colours and Pigments as Used by Artists*: the author lists a Cu–Cr pigment that “... may be prepared by several methods”.

Furthermore, the light-green enamel analyzed on shard #11 (Fig. 7b) proves to originate from the mixture of Sb and Cu. In particular the color is obtained by adding Cu

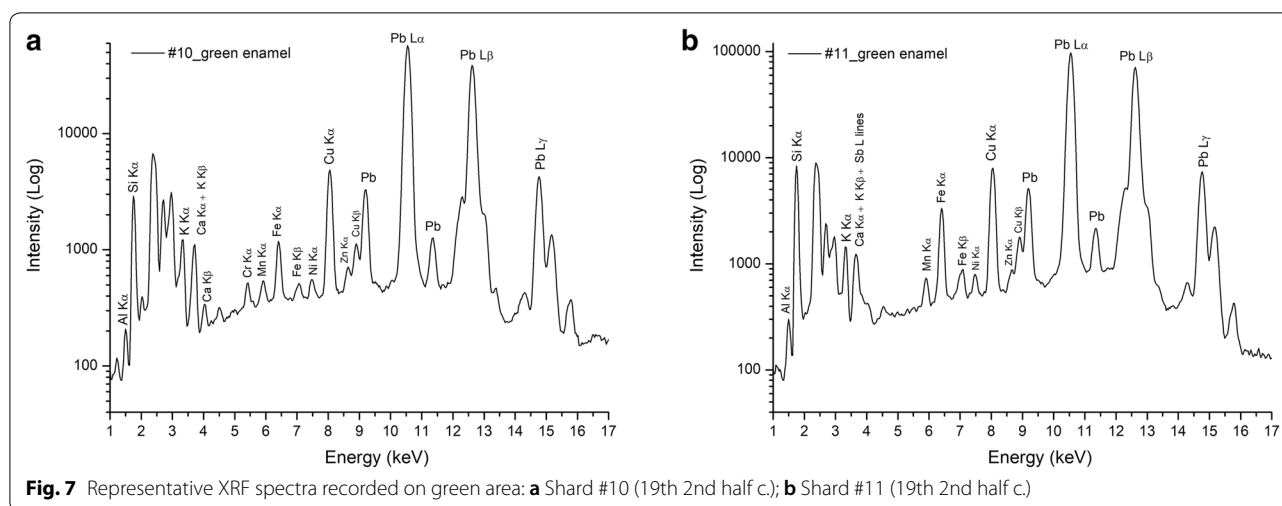


to the recipe for the Pb-Sb yellow detected on shard #7 (Fig. 3a). Noteworthy is to mention here the Renaissance recipe by Cipriano Piccolpasso, who listed in his *“Li Tre Libri dell’Arte del Vasaio”* (The Three Books of the Potter’s Art) in the mid-sixteenth century, a “mixed” green pigment consisting of a mixture of Cu, Pb and lead antimonate yellow [36]. Such record well traces the Italian origin of the later-developed northern European recipes that spread in the Far East in the nineteenth century as here demonstrated.

Naples yellow pigment: a tracer of Europe-Japan technological exchanges

Preparation procedures

As previously presented, the Pb-Sb yellow enamel detected on shard #7 (Fig. 3a) and the Pb-Fe-Sb-(Zn) yellow detected on shard #9 (Fig. 3b) both bear a Pb-Si-K matrix. This enamel composition is perfectly consistent with the recipe reported by Takamatsu in 1878 [31], therefore the preparation of the Pb-Sb and Pb-Fe-Sb-(Zn) compounds clearly differs from the calcination process required for the production of the European Renaissance Naples yellow. Moreover, no K was detected on the *mukozuke* dish (Fig. 3c) as expected for a lead antimonate compound prepared according to Valerio



Mariani's recipe for Giallo de' Vasari (Potters' yellow) published in the treatise *Della Miniatura* (1620) [18]. The implication is clear: the composition of the Japanese yellow enamels differ dramatically from the common Italian Renaissance recipes. The Renaissance yellow bears the fluxing agent NaCl, while the nineteenth-century Japanese yellows show the employment of a K-based flux, thus proving to belong to the group of contemporary antimony-based yellows developed in northern Europe [26, 37] on the basis of the original Italian process published by Passeri in 1758. The French scientist Fougeroux de Bondaroy (1769) was the first to have modified the process by adding potassium alum, and the other northern European recipes followed suit [26, 37].

The results perfectly fit into the trade picture of the nineteenth century, when new materials were being imported into Japan from northern Europe by the Dutch [4, 31, 38]. As a consequence of the isolation policy enforced by Tokugawa Shogunate, European materials available for porcelain production and paintings differed over the centuries. The *mukozuke* dish was decorated with a Renaissance yellow enamel of Italian origin imported by the Jesuits in the early 1600s, while the nineteenth-century porcelains were enameled with a yellow color based instead on the recipes developed in northern Europe in the nineteenth century.

Yet the Japanese created their own path and changed the actual production process of the coloring agent. European recipes required the calcination of a mixture of antimony oxide and lead oxide to achieve a stable Naples Yellow compound to be embedded in the NaCl fluxed matrix [16, 18, 39], as consistently detected on the *mukozuke* dish. The Japanese method involved instead the use of different bodies providing a Pb–Si–K matrix to which an antimony compound, pure antimony from Iyo

province [31], was then added. No previous calcination was carried out, thus leading to different enamel characteristics in terms of formed phases and reagents stoichiometry. Raman spectra (Fig. 2) provide further crucial information as to the firing temperatures of the antimony-based compounds, thus enabling the identification of their geographical area of origin and, along with XRF compositional data, their period of production.

The Raman signature of the pyrochlore solid solution

In order to fully understand this point, we have to consider that lead antimonate belongs to the family of compounds with the ideal pyrochlore structure having the general formula $A_2B_2O_6O'$ [13, 16, 27, 28, 40, 41]. B–O bonds are crucial to the cohesion of the crystal and a regular BO_6 polyhedron entails a more-distorted AO_8 polyhedron and vice versa [40, 41]. The (B–O) force constants are six to eight times stronger than those of $f(A-O)$ and $f(A-O')$ [40, 41]. Firing temperature, stoichiometric ratio, and small distortions of the pyrochlore structure strongly affect the Pb mode of the A_2O' lattice. Yet even a significant perturbation of the A_4O' network has a weak influence on the BO_6 network and overall rigidity of the structure [40, 41]. Therefore, from a vibrational standpoint, the two networks appear, on first approach, to be energetically independent [41]. The implication is clear: the firing temperature for enamel decoration affects the Raman wavenumber of the Pb mode to such a greater extent than the compositional base of the matrices in which the chromophore is embedded or the respective molar ratios. The lower the value of the diagnostic-peak wavenumber, the more stable the structure of the pigment owing to the increasing firing temperature [16]. It has been firmly demonstrated that as the firing temperature increases, the peak shifts to lower wavenumbers:

134 cm^{-1} at 800 °C, 130 cm^{-1} at 950 °C, and 124 cm^{-1} at 1100 °C [16]. The peak at 130 cm^{-1} observed on the Naples yellow used to decorate the *mukozuke* dish (Fig. 2c) matches the reported data perfectly, confirming an Italian Renaissance lead antimonate yellow fired at 900–1000 °C (*‘Feu de moufle’*). A firing temperature in the range of 900–1000 °C is incompatible with either the ceramic technology at Jingdezhen where overglaze enamels were fired at 700–800 °C [42, 43] or a Chinese enamel-on-metal technology with enamels fired at 600–700 °C [44, 45]. The latter, in particular, employs additional fluxing agents such as CaF_2 or even Bi_2O_3 [44] that lower the processing temperature, thus relying on a thermal-treatment duration which clearly differs between China and Europe in terms of kiln technology and heating rate.

The values of the diagnostic-peak wavenumbers observed on Chinese wares of any media, be it porcelain or metal, from any period and recipe, that were decorated with a lead antimonate yellow enamel [42, 44] fall within the range 136–142 cm^{-1} [42, 44]. They therefore fit into the firing-temperature range associated with both technologies in China, namely 600–800 °C [42–45]. As a consequence, a Chinese origin and firing of the yellow enamel is to be ruled out on the basis of such experimental evidences.

The strong peaks at 135 cm^{-1} and 141 cm^{-1} detected on the Japanese nineteenth-century Pb–Sb and Pb–Fe–Sb–(Zn) yellows (Fig. 2a,b), unequivocally point to a firing temperature in the range of 700–800 °C [16, 42] as confirmed by kiln practice for the period in Arita.

To conclude, based on the evidence discussed so far, the implication is very clear: the firing temperatures and chemical compositions of the Chinese Naples Yellows [42–46], be it on metal or ceramic, and the Japanese nineteenth century Naples Yellow, all differ dramatically from the Naples yellow of European origin that was detected on the *mukozuke* dish fired in the late Renaissance period (1630s). The latter shows a Raman signature consistent with the firing temperature of 950 °C as identified identified by Impey for the firing of Arita wares in the early Edo period [47, 48], as well as the firing temperature needed in Renaissance Italy to obtain a brighter and richer antimony-based yellow color [16]. The results point to a firing process the *mukozuke* dish underwent in western Arita, most likely at the Yanbeta kiln, under the supervision of European missionaries in the 1630s, before the final expulsion of all missionaries in 1639.

Historical recipes and scientific evidence: the connections Provenance and use of antimony

Documentary evidence and recent studies confirm that antimony was imported into Japan during the seventeenth century [4, 31, 38, 48, 49]. In particular, a Dutch

record for the year 1690 mentions the import of antimony into Japan as specifically needed for the decoration of porcelain wares [48, 49]. This instance is a highly significant one as Dutch traders knew the pigments being exported very well, considering that those same pigments, that is, lead-antimony and lead-tin yellows, started being imported into China from Europe around the same period, after the glass workshop had been established in the Palace in 1695 as ordered by the Kangxi Emperor to Jesuit missionaries [8, 42, 46, 50, 51]. In fact, Teodorico Pedrini (a Lazarist), who arrived in China with the painter Matteo Ripa to work at the court of the Emperor Kangxi, petitioned Rome in 1711 for the supply of antimony (*antimonio non preparato*) and enamels (*smalti*) needed for the production of new colors at the imperial workshop [8]. The missionaries were focusing on the preparation of new enamels, such as lead-tin yellow, lead-antimony yellow, arsenic-white and pink, that would enrich the Ming ceramic palette based on the traditional transition metals that Jingdezhen potters had been using for porcelain decoration until the Qing dynasty [8, 42, 46, 50, 51]. Moreover, the Dutch record mentioning antimony in 1690 triggered the suggestion [48] that antimony may have been the coloring agent used by Kakiemon in the late seventeenth century to produce the classic wares that took Europe by storm.

From a compositional standpoint, the chemical elements detected on the lead-iron-antimony yellow on shard #9 (Fig. 3b) offer, for the first time, crucial information regarding the actual recipe that formed the basis of Japanese porcelain decoration in the nineteenth and twentieth century. The presence of Fe, K, Zn, Sb and Pb strongly points to the recipe developed by Louis-Alphonse Salvétat at Sèvres in the mid-nineteen century. A Fe-based enamel characterized by the addition of Sb and Zn is listed in *Leçons de Céramique* published by Salvétat in 1857: “Fondant 880, Fleurs de zinc 35, Oxyde de fer hydraté jaune 70, Antimoine diaphorétique 15”.

Noteworthy is that the French recipe well matches the chemical composition of the yellow enamel detected on shard #9: the Arita enamel is a traditional Fe-based yellow with the addition of Sb, and Zn in a Pb–Si–K based matrix (Shiratama and other bodies as described by Takamatsu).

The Raman spectrum (Fig. 2b) has confirmed the nature of the compound acting as coloring agent, and has also shown the different stoichiometry of the reagents and the different firing temperature as compared to European kiln practice.

The significant presence of Zn is clearly the result of the influence of Salvétat’s method. Its addition, as also described by Theodore Deck in *La Faïence* published in 1887, helped obtaining different shades of an opaque

yellow in combination with Sb and Pb. Salvétat further explains in *Leçons de Céramique* how the use of Zn helps achieving a brighter yellow. Japanese potters appear to have followed such instructions to obtain a bright and opaque yellow for porcelain decoration.

This new enamel also reveals a very interesting transition from the Japanese traditional Fe-based yellow to the new Sb-Zn-containing European recipe. A clear instance of this change is represented by the Nabseshima shard (1690–1730) (Fig. 8a, b) and the *Ko-imari* Cup (1770–1790) (Fig. 8c, d) as compared to the shards analyzed in this work. The translucent Pb–Fe-based yellow enamel employed on the highly traditional Nabeshima shard and Ko-Imari cup appears to have been consistently used from the mid-seventeenth century, after the Jesuits had all been expelled from Japan, and throughout the eighteenth century. Japanese potters' practice of yellow enamelling with the translucent iron-based color in the eighteenth century is also reported in the book of recipes attributed to Johann Gregorius Höroldt (director of the Meissen manufacture) and dated to 24 December 1731: the "*Yellow in the Japanese manner*" is listed as consisting of iron compounds, contrary to the opaque Meissen yellow enamel based on lead antimonate [52].

Basically, Japanese potters ended up modifying their traditional translucent Fe-based recipe with minor additions of Sb and Zn to make it brighter and opaque. It appears very clear now how the transition must have been gradual and possibly smooth in terms of technological innovation as with other forms of applied arts. The same kind of approach emerges from *ukiyo-e* prints, where the incorporation of new synthetic colors of European origin proved gradual, selective, and coexisted with Japanese traditional practice [53].

The technological implications are very significant: the Renaissance opaque yellow enamel was based on a completely new and unknown technology that required Jesuit supervision in the first phase of enamel development in Japan in the 1630s, while the nineteenth-century enamels were obtained by modifying the European methods on the basis of the Japanese established local tradition. The Japanese way developed by adapting European recipes to Japanese practice. It appears pretty clear how such approach led to the modification of the European recipes: the lead-antimonate yellow became the result of a mixture of compounds with no previous preparation by calcination; and the innovative incorporation of limited amounts of antimony and zinc into the traditional translucent Fe-based yellow helped making it brighter and opaque.

More details on the effect of the use of zinc in yellow enamels for porcelain decoration is revealed by Alexandre Brongniart in the treatise *Colouring and Decoration*

of Ceramic Ware, published in 1898, where the author says "*Oxide of zinc enters into the composition of greens, yellows, yellow-browns and blues...Oxide of zinc...develops qualities necessary for the beauty of certain colours*". The use of zinc as an opacifier belongs to the European tradition as also testified by Salvétat in *Leçons de Céramique*.

European materials for painting and ceramics

The continued but very limited availability and circulation of European lead-based yellow chromophores in Japan is further clarified by the surviving early-seventeenth-century painting depicting *The Madonna with the Infant Jesus and her Fifteen Mysteries*: the pigment lead-tin yellow, unknown in Japan, was detected on the central inscription in Latin characters (*romaji*) "LOWADO SEIA O SANCTISSO SACRAMETO" [5]. This specific pigment belongs to the same family of Naples Yellow, the lead-antimonate enamel detected on the famous *mukozuke* dish fired in the *Kan'ei* period (1624–1644).

Noteworthy is to mention here that along with imported pigments and Japanese traditional coloring agents, flaxseed oil, a vegetable drying oil of European origin, unknown in Japan, not only was detected on the painting *The Madonna with the Infant Jesus and her Fifteen Mysteries*, but also on the western style screen *Map of the World and Famous Cities* (early seventeenth century), as the binding medium [5, 54]. This crucial instance has led Oka to emphasize the influence of Western technology on painting production, and to identify the Jesuit Seminario as the source of the imported materials [54].

Such unrecorded use of Western materials is perfectly consistent with a period of strict enforcement of anti-Christian edicts by the Tokugawa Shogunate [3, 12], with no chance for any written records mentioning any direct interaction with the missionaries to survive. Unfortunately, the yellow color on the screen *Map of the World and Famous Cities* was not analyzed.

Porcelain and paintings: one coloring agent for both productions

Additional important confirmation comes from the Yan-beta shard previously studied (Fig. 9) [4]: the composition of the green enamel analyzed on the shard perfectly matches the green pigment detected on the early-seventeenth-century screen *Western Kings on horseback* [55]. In terms of technological implications, an early use of a Cu–As–Zn based green pigment has been clearly detected on the works by the sixteenth-century Italian painter Stefano Sparano [56]. Once again, the practice of using the same materials for paintings and ceramics is of European origin and does not belong to Japanese tradition.

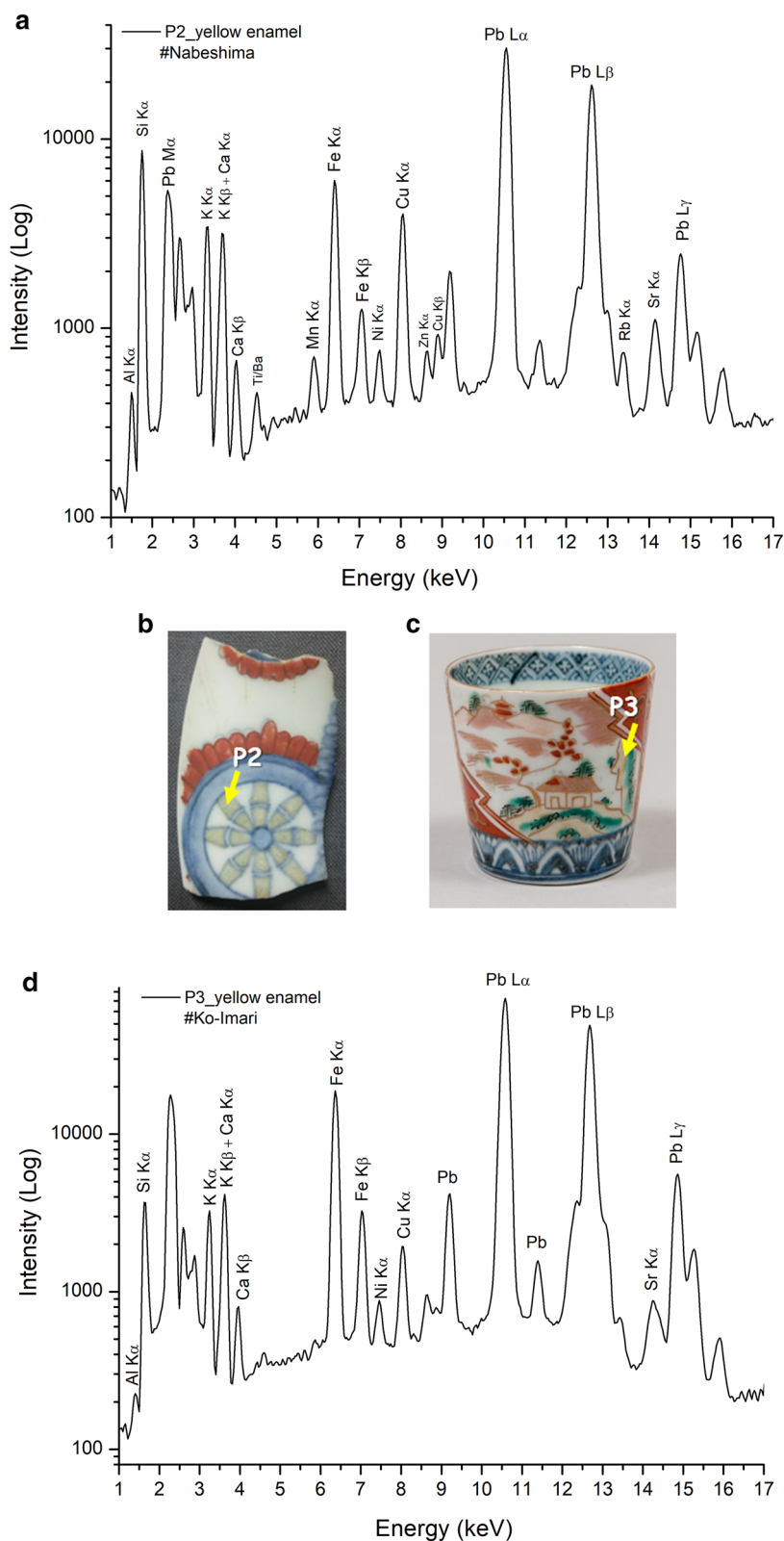
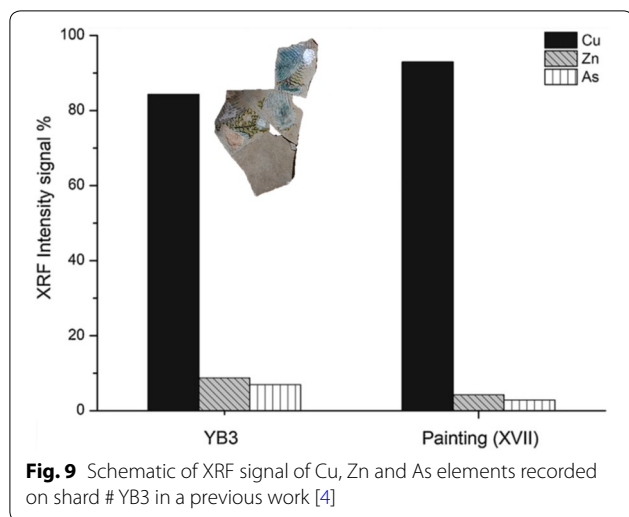


Fig. 8 **a, b** Nabeshima Shard (1690–1730) and Pb–Fe yellow enamel XRF spectrum; **c, d** Ko-Imari cup (1770–1790) and Pb–Fe yellow enamel XRF spectrum



Persecutions and closure of the country

European technology unknown to the Japanese was still available during the fierce persecutions that started in 1614, but on a very limited basis. Therefore, the overall analytical evidence confirms that the imported materials influenced color decoration on paintings and ceramics until the final expulsion of all Christians in 1639. The *sakoku* (closed country) policy established by the Tokugawa Shogunate led to the strict control of anything that entered the country, and trade with Europe ended up being monopolized by the Dutch and the Chinese in Dejima, thus ending the phase of spontaneous westernization of the applied arts that had started in the mid-16th century thorough the effort of Jesuits missionaries.

Blue coloring agents

Smalt: provenance and use

Of particular interest is also the case of the overglaze blue enamel, the glass rich in potassium and colored by cobalt oxide, used in Europe since the fourteenth century for paintings and ceramics alike [57–60]. This blue pigment, also named smalt (*saffre* in French, *smalto* in Italian), has been detected on a hand-colored Japanese Buddhist print dated to the late 16th century [61], and on *ukiyo-e* paintings from the seventeenth to the late eighteenth century [38]. Recent systematic studies have demonstrated that smalt had initially been imported into Japan by the Jesuits for porcelain decoration in the late Kan'ei period (1624–1644), thus resolving the long-standing issue of the origin of the material [4, 62]. Afterwards the Dutch monopolized the trade [38]. From the *Kanbun* era (1661–1673) the imported material was also employed on *ukiyo-e* paintings [38]. Noteworthy is that there are no earlier examples of the use of smalt in the Asian paper or silk

painting tradition [38], so Japan appears to have been the first country where the material started to be used under the supervision of Jesuit missionaries in the late sixteenth century.

This pioneer use of imported European materials proves perfectly consistent with the detection of lead-antimonate yellow on the rare *mukozuke* dish (Fig. 2c, 3c), of lead-tin yellow on the painting *The Madonna with the Infant Jesus and her Fifteen Mysteries*, and of flaxseed oil on both *The Madonna with the Infant Jesus and her Fifteen Mysteries* and the western style screen *Map of the World and Famous Cities*. No wonder we find smalt on *ukiyo-e* works from the latter half of the seventeenth century: the supply of the material, as for the case of porcelain production [4], had stabilized at the time by means of the trade with the Dutch, who, to cope with the high demand, established a production facility in Holland [4, 38]. The pigment innovation of the late 16th century, as testified by the hand-colored Buddhist print [61], kept showing strong roots in the artists' creative approach. The detection of smalt on *ukiyo-e* paintings is very significant as *ukiyo-e* represented a new form of art that shaped popular culture during the Tokugawa Shogunate. Once again, as for the craze for anything European (*Namban*) that took Japan by storm in the Momoyama period (1568–1603), European innovations kept inspiring new experimentation that shaped the artistic forms of the whole Edo period (1603–1868).

Paintings and enameled porcelains were expensive commodities; therefore it makes perfect sense to have found the imported materials on works reserved for the wealthy. Significant instances of the use of smalt by Japanese masters come from the seventeenth-century painting *Flowers and Rocks* by Ogata Korin (1655–1716) and the late-eighteenth-century *Moonlight Revellry at Sagami Dozo* by Kitagawa Utamaro [62].

In terms of pigment origin, full confirmation comes from the blue color analyzed on a Japanese votive painting dated 1682 [63]: the chemical composition of the pigment is Co–Ni–Fe–As, which, as expected, perfectly matches the composition of the European smalt-based enamel imported into Japan by the Jesuits in the late 1630s for overglaze blue decoration of porcelain wares [4]. It is worth noting here that the practice of using the same coloring agents for paintings and ceramics belongs to the European technological tradition. Japan therefore established itself as the first country to have incorporated European Renaissance technology into traditional Far Eastern applied arts.

Smalt, as for the case of Naples Yellow, lead-tin yellow and arsenic-opacified white, will acquire a central role in the new opaque enamel palette developed at

Jingdezhen (China) from 1720s for the decoration of imperial wares [7, 8, 42, 64, 65].

Foreign coloring agents in the late Edo period (eighteenth and nineteenth centuries)

Ukiyo-e paintings were gradually replaced by woodblock prints, an inexpensive mass produced form of art. Publishers needed to switch to less expensive and ready-to-use materials in order for the prints to be enjoyed by the public at large: Prussian Blue, a synthetic pigment developed in the Old Continent in the early eighteenth century [66], started to replace smalt and coexist with native natural pigments from the early nineteenth century [67], as testified by masterpieces such as *Doshoku Sai-e* by the painter Ito Jakuchu (1716–1800) [68]. It is interesting to note here that smalt and Prussian Blue have been identified as being part of the *Kaishunsai* paints, the pigments collected by the Nabeshima family in the early nineteenth century and stored in Takeo city, Saga prefecture [69]. Takeo city is located in the vicinity of Arita, where the production of blue-decorated porcelains was recently discovered to have developed in the late 1630s on the basis of a smalt-based enamel imported by the Jesuits [4]. The Nabeshima family controlled porcelain production in Arita, and the clan's crucial role became evident on the occasion of the organization and unification of the kilns that occurred in 1637 [70]. Such change was needed to exploit the full potential of the porcelain trade. So the presence of the pigments in the collection further testifies to the influence of European technology on art production in Japan throughout the Edo period.

Comparison with China: smalt and European chromophores

European technological influence on Chinese arts followed the exact same path established in Japan one century earlier. The Chinese, after the first technological exchange with the Jesuits in 1695 [7], started incorporating new imported materials into their artistic practice: smalt, lead–tin yellow, lead–antimonate yellow, Pb–Sb–Sn yellow, arsenic white, Prussian Blue and new artificial pigments saw an extensive use in ceramics and paintings throughout the Qing dynasty and up to the present day [42, 64, 65, 71]. Contrary to the case of Japan, Jesuit missionaries enjoyed imperial patronage in China and were free to exert their influence on the production of polychrome wares at Jingdezhen in the late seventeenth and early eighteenth century. In Japan the Jesuits were forced to hide when the young porcelain industry in Arita started developing the first overglaze enamels. Therefore the same gradual modification and incorporation of European recipes as identified in eighteenth-century China and late-nineteenth-century Arita, would have

been impossible in the early seventeenth century when the fierce persecutions culminated in the final expulsion from Japan of all missionaries in 1639.

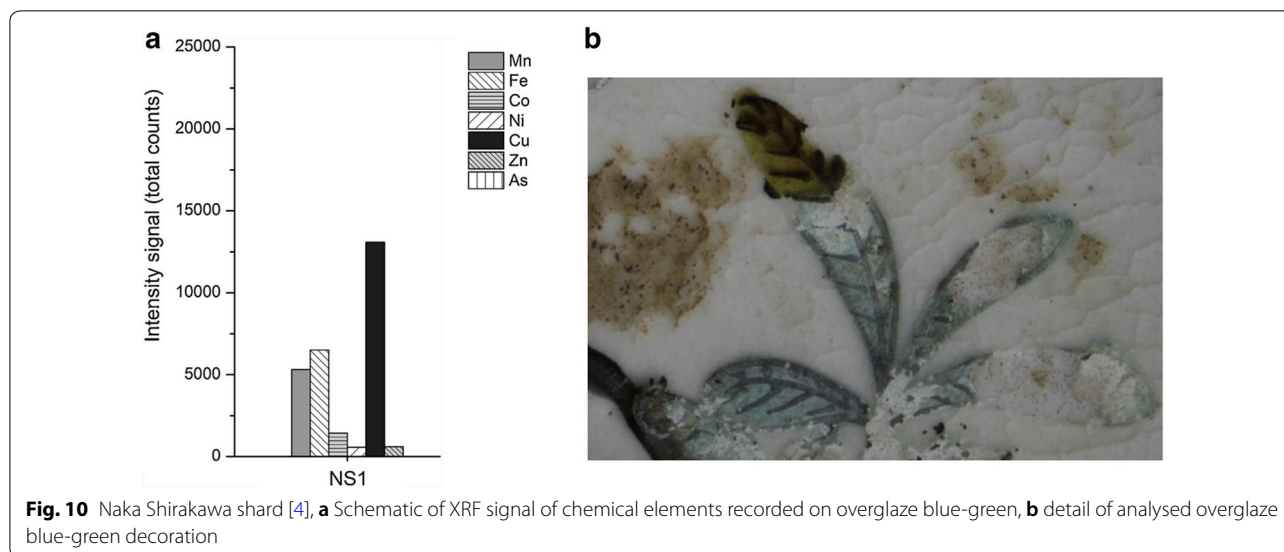
The two painting and ceramic traditions, Japanese and Chinese, however, differed for the extensive use of smalt that characterized the former and that almost completely lacked in the latter [7, 47, 72–75].

European recipes for the global market: production innovations in the porcelain industry of Arita

Domestic consumption of porcelain in Japan had spread by the late eighteenth century [76], yet the porcelain industry started to face a deep decline due to the reduced trade with Europe, kiln destructions by fire [76], and the fierce competition by the mass-produced Chinese copies of Imari wares that had flooded the European market in the eighteenth century [73, 74]. Chinese traders were capable of taking full advantage of any business opportunity arising in the Far East. In this highly competitive environment, kiln owners in Arita needed to devise new strategies to make the whole process less expensive and meet domestic and western taste. The development of new colors involved the use of synthetic pigments made in Europe: the present results show how such decision became a dominant factor in enamel production. A clear shift to the newly-developed and cheaper materials took place in the nineteenth century, after a predominance of the traditional coloring agents in the seventeenth and eighteenth centuries (transition metals), yet with the continued import of the smalt-based blue enamel from the Old Continent.

In particular Takamatsu describes the imported blue material from Europe in 1878 [31] by saying: “No certain information of the manufacture of this substance has been obtained at present. It is said that it may be prepared by fusing crude oxide of cobalt with a mixture of clay and alkalies, so as to form a kind of blue glass, or smalt. At present foreign smalt is largely used, being sold at a cheaper rate”.

Takamatsu therefore records that the blue pigment, known in Japan as *Hanakonjo*, was being imported as a ready-to-use material, thus confirming that Japanese painters and potters had not yet established a local production and kept relying on the European supply until the early twentieth century [31, 77], as also demonstrated in this work. The continued use of imported smalt remained unchanged since the late 16th century, as testified by the Japanese votive pictures and *ukiyo-e* paintings, up until the twentieth century [77]. Our results have also shown that the blue pigment employed in Arita from the late nineteenth century consisted of the newly developed Co-based synthetic material of European origin that gradually replaced smalt for overglaze decoration,



and low-grade natural-cobalt-ores for underglaze decoration (Fig. 5a, 6a). These findings, along with previous studies [3, 4], fully reveal that even though the Japanese had been exposed to European materials from the late 16th century, they didn't succeed in mastering the technology needed to reproduce them until the early twentieth century. This further explains why the very limited use of the scarce and expensive lead-based yellows during the Momoyama and early Edo periods suddenly stopped after the final expulsion of all Christians and Jesuits in 1639. The Japanese were still in the earliest stage of enamel-firing experimentation and therefore needed a new chromophore to replace the unavailable Jesuit-related yellows (lead antimonate and lead-tin yellows). Yellow was the most difficult color to fix for Arita potters [70] and the issue was resolved by means of the cheaper and easier-to-fire iron yellow, likely through Chinese supervision. But the practice of replacing imported European materials of unknown technology with Chinese pigments could not be successfully carried out in the case of smalt. The Japanese were not capable of reproducing the same coloring effect with any of the materials available to them at the time, locally or from China. This is firmly demonstrated by a previous study by Montanari et al. [4]: the polychrome decorated shard (1640–1650) excavated at the Naka-Shirakawa kiln-site (Fig. 10a, b) reveals the attempt to use a low-grade cobalt ore imported from China (commonly employed for underglaze blue decoration) to achieve an overglaze blue-green effect. The attempt failed as the low-grade cobalt ore was unsuitable for overglaze decoration. It becomes undoubtedly clear that as soon as the Jesuits left the country in 1639, the technology to obtain smalt could not be reproduced,

and the failed Naka Shirakawa attempt testifies to the tireless effort by Arita potters to find a way of replacing the imported blue material. Potters could not succeed in that task, and were therefore forced to keep relying on imports from Europe. Furthermore, the Naka-Shirakawa shard bears a Fe-based yellow enamel. The presence of both the 'experimentally-failed' overglaze blue-green enamel and the Fe-based yellow enamel on the same shard testifies to the frantic situation Arita potters were facing in the earliest stage of polychrome porcelain production right after Europeans had been expelled from the country in 1639. The shard is offering a glimpse of the period that followed immediately after the total closure of the country around the early 1640s: Japanese potters had managed to replace the Jesuit-related yellow materials with the iron yellow, but could not replace the smalt needed for overglaze blue decoration as it wasn't available in China at the time. Therefore they had to rely entirely on imports by the Dutch, the only Europeans allowed to trade with Japan during the Tokugawa Shogunate. Trade of ceramic materials and development of production techniques became the major factors impacting the competition between potters. The profitable market for enameled wares became the aim of Arita kilns as well as their fierce battleground.

To conclude this part, the results demonstrate that in the late Meiji period (1868–1912), as soon as European presence became relevant once again in the country, contrary to the Edo period, Japanese potters finally managed, for the first time, to establish their own production of antimony yellow and synthetic blue materials. The main difference between the closed country policy of the Edo period and the Meiji policy of strong westernization is

that the latter led to the establishment of a full-fledged, state-of-the-art industry that enabled Japan to become, once again after 400 years, the hub for European materials and technology in the Far East.

Ready-to-use pigments in the twentieth century: from Europe to Japan and China

The scientific evidences also confirm the suggestions by Kerr, Wood and Watt [7, 43, 71] regarding the replacement of Jingdezhen overglaze and underglaze materials with newly developed and ready-to-use pigments imported from France, Germany and Japan in the early twentieth century. In particular, Watt reports the practice at Jingdezhen of mixing natural low-grade Chinese cobalt ores with newly developed synthetic blue pigments in the early twentieth century [71]. The technology transfer appears very clear on the basis of our results: shard #7 (Fig. 5a) reveals that such practice started in Arita in the late nineteenth century and then spread to Jingdezhen, when the new Japanese-made pigments became an important part of the trade with China in the early twentieth century. Further confirmation comes from the Sb–Cu green enamel detected on shard #11 (Fig. 7b).

Takamatsu also describes the preparation of blue enamels by listing the reagents involved in the process: he basically tells us that the imported ready-to-use material *Hanakonjo* was embedded into a Pb–Si–K matrix (a mixture of *Shiratama*, *Tonotsuchi* and *Hinookaseki*) [31]. This further confirms that the Japanese practice of using imported blue pigments from Europe for paintings and ceramics had not changed since the first encounter with the Jesuits in the late sixteenth century. The results presented for shard #9 and a modern high artistic value cup further prove this continued practice (Fig. 6a, b).

Shard #9 therefore offers a clear instance of the shift from the smalt-based recipe to the newly developed Co-based synthetic ones: the XRF spectrum shows a compound of Co and Zn (Fig. 7a). Such composition is in perfect agreement with the recipe published in 1869 by Thomas Salter in *Field's Chromatography or Treatise on Colours and Pigments as Used by Artists* [66]. The author lists a *Zinc-Cobalt Blue* saying that "... For tinting porcelain...it is admirably adapted, imparting thereto a very pure dark blue of extraordinary beauty". Charles-Ernest Guignet further describes in *La céramique ancienne et moderne* (1899) the effects that the incorporation of Zn in the blue recipe allows to achieve: "... For sky blues, add zinc oxide...so that the mixture takes on a tone closer to pure blue".

Conclusions

The analytic results reported here demonstrate how the study of ceramic enamels may provide valuable information not only on the history of porcelain production, but also on materials employed in Asian painting and printing traditions. We have demonstrated how the long-thought exclusive Chinese influence on Japan, especially on ceramics and paintings, was only one aspect of the Japanese technological development of coloring agents. The arrival of Europeans in 1543 triggered a privileged exchange that enabled Japan to become the first hub of European technology in the Far East, thus reversing, for the first time, the centuries-old predominance of China as the main actor in the spread of western culture throughout Asia. The new evidence confirms that the pattern of influence established by the Jesuits in the late 16th century laid the foundation for a new and independent Japanese cultural development. Such development will strongly influence China in the late nineteenth and twentieth centuries. Extensive experimentation with new synthetic pigments and enamels imported from the Old Continent will mark this extraordinary process of Japanese innovation. However, the two traditions will be distinguished by the extensive use of smalt in Japan as opposed to its very limited use in Chinese paintings, as well as in Chinese ceramic production before the eighteenth century.

The new evidences reveal that synthetic pigments started to be produced in Japan in the twentieth century on the basis of European original recipes. Such pigments will also be exported to China for porcelain decoration. Japan, as occurred four centuries earlier with the Jesuits, became once again the hub for western material circulation in the Far East.

Moreover, Raman results have demonstrated, for the first time on Far-Eastern porcelains, that the shift of the Pb mode of the A_2O' lattice of lead antimonate yellow (Naples Yellow) provides important information on the firing temperature for enamel decoration. Such characteristic, along with its chemical composition, enables the identification of the area of origin and period of manufacture of the yellow enamel.

Abbreviations

EDXRF: Energy-dispersive X-ray fluorescence; XRF: X-ray fluorescence.

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Authors' contributions

The research project was conceived by RM, who also prepared the manuscript. The shards were selected by NM and RM. The analyses were carried out by RM, MFA, SS. The results were interpreted by RM, MFA, PhC, and CP. PhC and CP also revised the manuscript. All authors read and approved the final manuscript.

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