

X-ray Fluorescence Spectroscopy Study for Tracing Metal Elements on the World's Oldest Metal-Type Printed Book from the Goryeo (高麗) Dynasty of Korea in 1239, *The Song of Enlightenment* (南明泉和尚頌證道歌)

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Abstract

In a groundbreaking study using X-ray fluorescence (XRF) spectroscopy, an ancient book, believed to have been printed in 1239 during Korea's Goryeo Dynasty, was examined to uncover the printing technique behind its creation. The book in question is a rare *Gongin version* (空印本) of *The Song of Enlightenment* (南明泉和尚頌證道歌), a text that holds immense historical significance and was designated as a Korean Treasure in 2012. While the colophon of the book explicitly claims it was printed using movable metal type in 1239, a contentious debate exists among scholars on how it was printed. Some interpret the colophon differently, suggesting the book was produced using duplicated woodblocks—a common printing method of the time. To resolve this mystery, a portable XRF device was employed to detect the presence of metal elements that could confirm the use of metal type. The results were compelling. Metal elements were found concentrated in and around the printed characters, with iron (Fe) levels showing a striking contrast: printed characters, inked with traditional Korean ink (*meog*, 墨), exhibited iron (Fe) content as high as 1,530 ppm—nearly twice the concentration found in the surrounding blank paper (around 800 ppm). More intriguingly, sulfur (S) content varied dramatically, ranging from 2,300 ppm to a remarkable 13,000 ppm, suggesting a chemical reaction linked to the ink or the printing process. Copper (Cu) and antimony (Sb) were also detected, though in smaller quantities between 20 ppm and 90 ppm for copper, and a consistent 300 ± 50 ppm for antimony. Given that higher levels of iron (Fe) and sulfur (S) were consistently found in the printed characters across all measured pages, the evidence strongly points to the use of metal type, not duplicated woodblocks, in the production of this version of *The Song of Enlightenment*. This quantitative XRF analysis not only casts light on the printing technique of this particular book, but also offers a reliable method for analyzing similar ancient texts, helping to unravel the mysteries of early printing practices in Korean history.

Keywords: the world's oldest movable metal type printed book, *The Song of Enlightenment* (南明泉和尚頌證道歌), movable metal type, X-ray fluorescence (XRF), metal element detection, printing method

1. Introduction

In 2021, a collaborative international research initiative titled “From Jikji to Gutenberg” was launched to examine the Buddhist scripture *Jikji Simche Yojeol* (直指心體要節), commonly known as *Jikji* (直指), a key text printed using movable metal type during Korea's Goryeo dynasty (高麗, 918–1392). The project, supported by a grant from the National Endowment for the Humanities (NEH), is spearheaded by the UNESCO International Centre for Documentary Heritage (ICDH) and the University of Utah's J. Willard Marriott Library ([The University of Utah 2025](#); [The Korea Times 2023](#)). It involves over 40 scholars across 13 time zones, collaborating with major institutions such as the Library of Congress, the Smithsonian Institution and the Gutenberg Museum in Germany. The primary goal of the project is to explore *Jikji* and the *Gutenberg Bible*, two pivotal cultural artifacts that are widely regarded as foundational to the development of printing in Eastern Asia and Western Europe, respectively. Both works were designated as part of UNESCO's Memory of the World Register in 2001. However, in the West, the fact that *Jikji* predates the Gutenberg Bible by 78 years remains largely unacknowledged.

Jikji was published in 1377 during the Goryeo dynasty at Heungdeok Temple (興德寺) in Cheongju (淸州), Korea. This *makes Jikji* the world's oldest extant book printed with movable metal type, currently recognized by UNESCO, 78 years earlier than the *42-line Gutenberg Bible* (1455), which is often cited as the earliest European book printed with mass-produced movable metal type ([The University of Utah 2025](#); [The Korea Times 2023](#); [UNESCO 2025](#); [UNESCO 2025b](#); [BnF 2025a](#)). The only surviving original copy of *Jikji* is currently held at the National Library of France in Paris ([UNESCO 2025a](#); [BnF 2025a](#)). It was recently displayed publicly for the first time in fifty years as part of the special exhibition *Imprimer! L'Europe de Gutenberg* (April 12 to July 16, 2023) ([BnF 2025b](#)). *Jikji* is also officially recognized by Guinness World Records as the oldest extant book printed with movable metal type, further cementing its significance in the history of printing ([Guinness World Records 2025a](#)).

The history of movable metal type printing in Korea, however, stretches back even further. The first documented use of movable metal type dates to 1234, during the Goryeo dynasty. At that time, the Goryeo government commissioned a civil servant named Choi Yun-ui (崔允儀, 1102–1162) to print a new edition of *Sangjeong Gogeu Yemun* (詳定古今禮文, *The Prescribed Ritual Text of the Past and Present*), a Confucian text spanning 50 volumes. Carving woodblocks for each page would have been impractical, especially since skilled woodblock carvers were already occupied with the Tripitaka Koreana project, a monumental task under the Mongol invasion taking place around that time. As a result, Choi developed a method using reusable metal types, likely cast from bronze. Between 1234 and 1241, 28 copies of the *Sangjeong Gogeu Yemun* were printed, making this the history's earliest known use of metal movable type ([Chon 2025](#); [Guinness World Records 2025b](#); [Sohn 1959](#); [Sohn 1971](#)).

While Korea has a long history of using movable metal type, *Jikji* was not internationally acknowledged as the oldest extant book printed with movable metal type until 1972. This recognition sparked renewed interest in Korea, leading to efforts to find even earlier examples of books printed using this technology ([Park 1988](#); [Oh, 1988](#); [Park 2020a](#); [Park 2020b](#); [Yoo and](#)

Kim 2021). One text under close examination was the *Gongin version* (空印本) of *The Song of Enlightenment* (南明泉和尚頌證道歌), which, according to the colophon, was printed with movable metal type in 1239. Despite the early claim in the colophon, there was considerable debate among bibliographers in Korea for nearly 50 years, and *Gongin version* (空印本) was not recognized as a book printed with movable metal type until recently (Park 1988; Oh, 1988; Park 2020a; Park 2020b; Yoo and Kim 2021; Yoo 2022a; Yoo 2022b; Yoo 2022c; Yoo 2022d; Yoo 2022f; Yoo 2023; Yoo 2024; Yoo and Yun 2024).

Based on many evidence from the previous studies on *The Gongin version of The Song of Enlightenment* with very similar versions, Jikji and Gutenberg Bible, it has been accepted as the world oldest metal type printed book. The major evidences were detailed image comparisons, image analysis, colophon dates, font and printed character changes between versions, and ink tone characteristics (Yoo and Kim 2021; Yoo 2022a; Yoo 2022b; Yoo 2022c; Yoo 2022d; Yoo 2022f; Yoo 2023; Yoo 2024; Yoo and Yun 2024).

In January 2023, the American Printing History Association (APHA) incorporated new research, including image and ink tone analysis, confirming that the *Gongin version* of *The Song of Enlightenment* is the world's oldest extant book printed with movable metal type, dating to 1239 (American Printing History Association (APHA) 2023). This re-evaluation has pushed back the recognized origins of movable metal type printing, positioning the *Gongin version* of *The Song of Enlightenment*, as the earliest extant book printed with movable metal type printing technology.

Despite these new research results and changes in perception, there are still widespread claims that the *Gongin version* of *The Song of Enlightenment* was printed with duplicated woodblocks. To resolve the long-standing controversy on the *Gongin version*, researchers turned to X-ray fluorescence (XRF) spectroscopy; a powerful, non-destructive analytical technique widely used in the study of cultural heritage materials. Research on the compositional analysis of cultural heritage materials—including ink, minerals, pigments, paper, and bone—using X-ray fluorescence (XRF) is actively conducted worldwide (Wenz 2022; Edwards et al. 2022; SLAC 2025; Wagner et al. 2023; Obregón et al. 2021, ESRF 2025; Kobylarz et al. 2023; Ridolfi 2012; Manso et al. 2008; Gomes et al. 2024). Recently, at Stanford University's state-of-the-art Synchrotron Radiation Lightsource (SRL), non-destructive multispectral and XRF imaging was applied to detect metal elements from ancient Korean books printed with movable metal type and books printed with woodblocks (Wenz 2022). They reported that significant amounts of metal components were detected exclusively in books printed with movable metal type, providing researchers of the “From Jikji to Gutenberg” project with a solid scientific foundation (Wenz 2022).

One more evidence for metal type printing is strongly desired. To resolve the debate surrounding the origins of the *Gongin version* of *The Song of Enlightenment* can be done similarly by using XRF measurement.

2. Experiment

2.1 The Book: *Gongin version* of *The Song of Enlightenment*

Designated as a Korean Treasure in 2012, the *Gongin version* of *The Song of Enlightenment* was initially classified as a duplicated woodblock-printed text rather than one

produced with movable metal type (Korea Heritage Service 2012). Figure 1 shows its front cover and bookbinding method, which uses thread passed through seven drilled holes. Traditionally, Korean books are bound using a five-stitch method, with five holes drilled into the pages, while Chinese and Japanese books follow a four-stitch method with four holes. A key distinction is that Korean books typically have an odd number of binding holes, whereas Chinese and Japanese books use an even number. Despite its modest size (24.1 cm × 15.0 cm), the *Gongin* version is bound with a rare seven-stitch technique, suggesting efforts to reinforce its durability, possibly for use across multiple generations. This meticulous binding highlights the text's significance and the craftsmanship involved in its preservation. There are no signs of rebounding or repair for conservation. The original book binding and damage by normal wear and tear for almost 800 years can be seen.

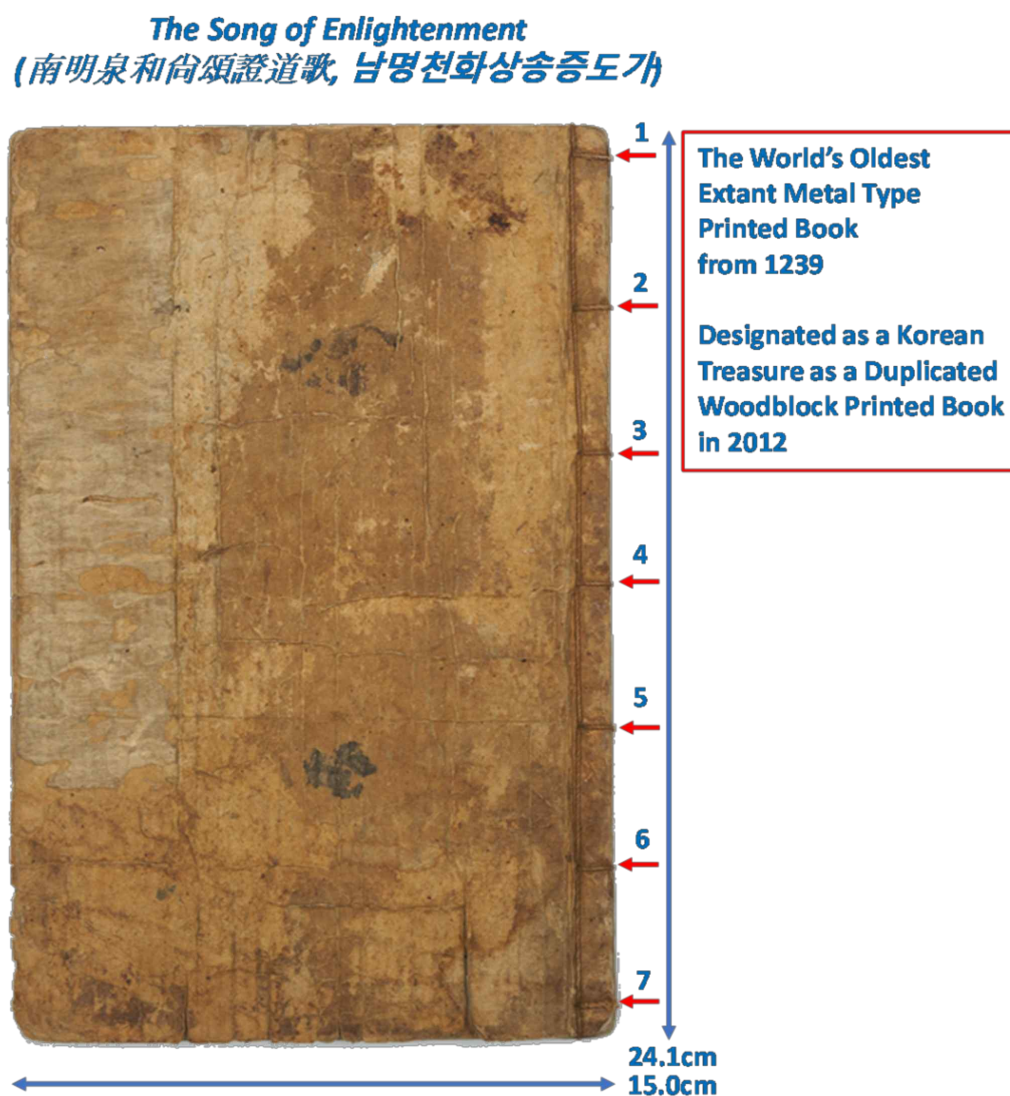


Figure 1. The *Gongin* version of *The Song of Enlightenment* printed using metal type in 1239 (book size: 24.1 cm (H) x 15.0 cm (W)).

Figure 2 presents the first printed page of the *Song of Enlightenment* next to the backside of the front cover. Like most traditional Korean books, the *Gongin version*, along with the closely related edition, *Samseong version* (三省本) (Korea Heritage Service 1984), which contains the same content, but was printed using woodblocks, follows the standard format of classical East Asian texts. Writing is arranged vertically from left to right, and the book is read by turning pages from left to right. As a result, the front cover is bound on the right-hand side, a convention shared by Chinese and Japanese books of the same period. Figure 3 shows the last printed page (the front side of 44th leaf) with the colophon stating the book was printed with movable metal type in early September 1239 (highlighted using blue and red rectangles) and the backside of 43rd leaf of the *Gongin version* of *The Song of Enlightenment*.

A closely related edition, the *Samseong version* (三省本), contains the same content but was printed using woodblocks. The *Samseong version* was designated as a Korean treasure in 1984 (Korea Heritage Service 1984). This version of the *Song of Enlightenment* is universally recognized as a woodblock-printed text, as its production method has been thoroughly verified. Clear evidence of woodblock printing is present in the form of characteristic imperfections, such as cracks following the natural grain patterns of the wood and missing strokes in characters caused by wear and tear from repeated use (Park 1988; Oh, 1988; Park 2020a; Park 2020b; Yoo and Kim 2021; Ok 2013; Cheon 1984; Cheon 1988). Since the Goryeo Dynasty, Korea has maintained a tradition of rapidly printing limited quantities of texts using movable metal type for initial distribution, followed by independent engraving of those texts into woodblocks for broader circulation. In many instances, books printed with movable metal type served as the master copies for creating duplicated woodblocks. Local governments and temples produced their own woodblocks, using them to print books as needed for regional distribution. Once a sufficient number of copies had been printed, these woodblocks were carefully preserved and could be reused whenever necessary. The durability of woodblocks allowed them to remain in use for centuries, as exemplified by the more than 80,000 wooden printing blocks of the Tripitaka Koreana engraved in the 13th century (UNESCO 2025c).

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The Song of Enlightenment
(南明泉和尚頌證道歌, 남명천화상송증도가)

Front Side of First Leaf

Backside of Cover Page

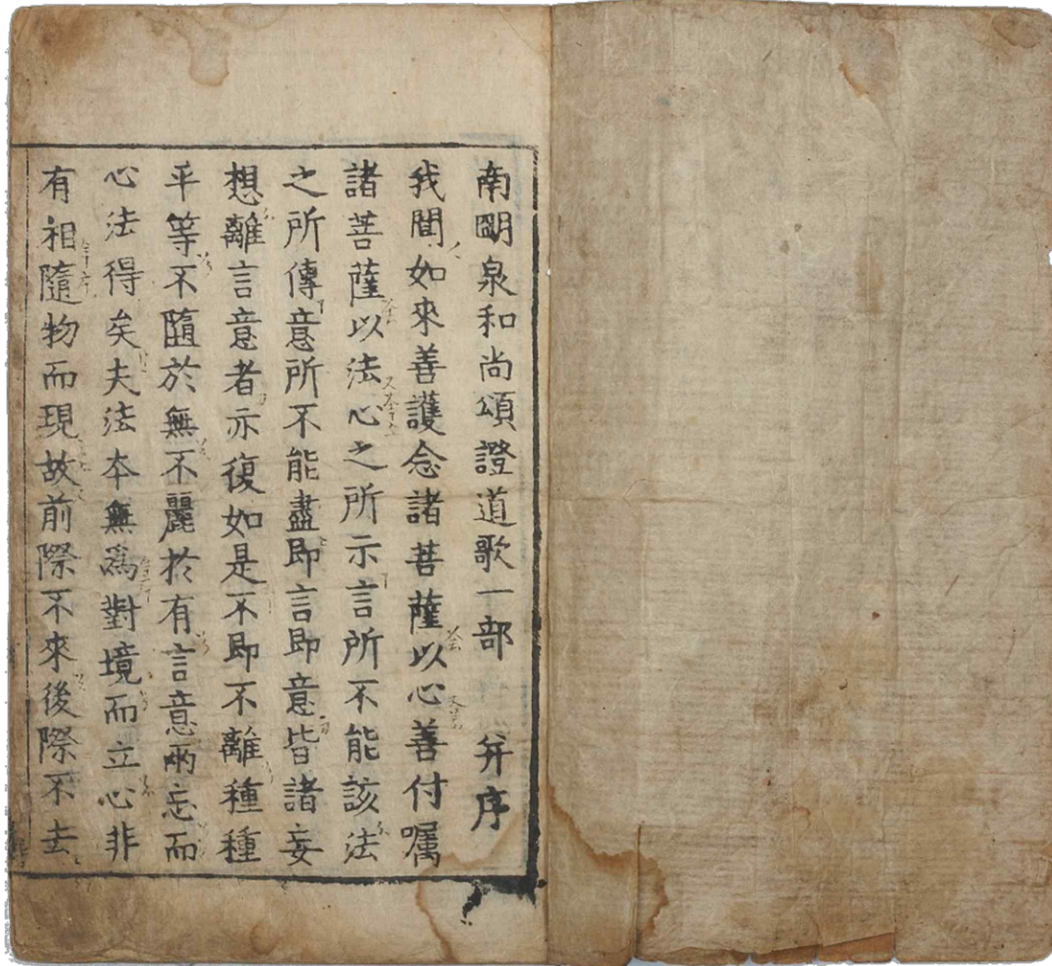


Figure 2. The first printed page (front side of folded first leaf) and the backside of front cover of the *Gongin* version of *The Song of Enlightenment* printed using metal type in 1239 (book size 24.1 cm (H) x 15.0 cm (W)).

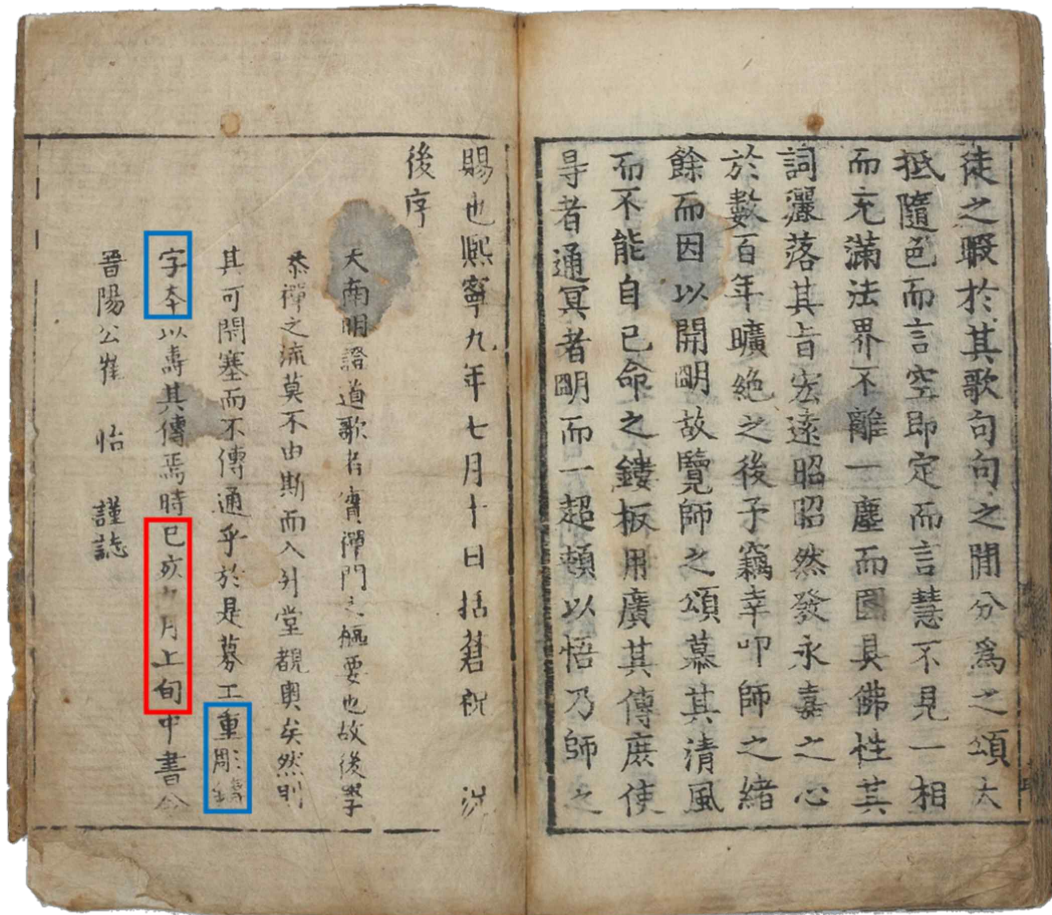
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The Song of Enlightenment (南明泉和尚頌證道歌, 남명천화상송증도가)

Last Printed Page with Colophon

Backside of 43rd Leaf



Printed using Metal Type

Early September 1239

Figure 3. The last printed page with colophon stating the book was printed using metal type in early September 1239 (highlighted using blue and red rectangles) and the backside of 43rd leaf of the Gongin version of *The Song of Enlightenment*.

The ongoing controversy surrounding the printing method of the *Gongin* version of *The Song of Enlightenment* has persisted since the 1970s and has been comprehensively summarized in previous works (Yoo and Kim 2021; Yoo 2022a; Yoo 2022b; Yoo 2022c; Yoo 2022d; Yoo 2022f; Yoo 2023; Yoo 2024; Yoo and Yun 2024). To avoid redundancy, this paper does not revisit the differences between the versions. Detailed information about other editions of *The Song of Enlightenment* has been extensively covered elsewhere (Park 1988; Oh, 1988; Park 2020a; Park 2020b; Yoo and Kim 2021; Yoo 2022a; Yoo 2022b; Yoo 2022c; Yoo 2022d; Yoo 2022f; Yoo 2023; Yoo 2024; Yoo and Yun 2024). This study focuses exclusively on presenting the XRF measurement results and their interpretation.

2.2 Portable X-ray Fluorescence Spectroscopy

XRF technology spans a broad range of equipment, from large-scale experimental facilities at Stanford University (Wenz 2022; Edwards et al. 2022; SLAC 2025) and the European Synchrotron Radiation Facility (ESRF) in Grenoble, France (ESRF 2025), to more accessible portable handheld devices (Kobylarz et al. 2023; Ridolfi 2012; Gomes et al. 2024; Evident 2025). While the fundamental principle remains the same across these systems, there are notable differences in spatial resolution and the detection limits for various elements. Ideally, equipment with high spatial and energy resolution would be used, but such advanced facilities are rare and can be challenging to access in practical settings.

In this study, a portable XRF (Olympus Evident Vanta C Series, Massachusetts, USA) was employed for elemental analysis (Evident 2025). The device allows for quantitative analysis of 26 elements, ranging from magnesium (Mg) to uranium (U), excluding noble gases such as helium (He), neon (Ne), argon (Ar), and krypton (Kr). Due to the inherent measurement principles of XRF, elements lighter than magnesium (Mg) and noble gases cannot be separated or quantified. For elements lighter than sodium (Na), the total content within the measurement area (more precisely, the measurement volume) is reported as LE (lighter element) values.

It is equipped with a silicon drift detector and a 40 keV X-ray tube with a rhodium (Rh) anode. It is designed and ideal for scrap sorting, PMI (Positive Material Identification), geochemistry, mining, jewelry ID, soil analysis and an array of other applications. Among many operation modes, Geological & Environmental Modes - Geochemistry (Geo / Mining) were used. 4-watt X-ray tube with application-optimized anode material, rhodium (Rh), was used. Two 20 s scans at 10 keV and 40 keV were performed per measurement point. Prior to the actual measurement, factory recommended daily calibration using a 316 Stainless Steel (SS) calibration disk provided by manufacturer. To ensure the distance between X-ray source and testing material the XRF analyzer nose flush was placed against the testing material (calibration disk or *Hanji* paper).

The ancient *Gongin* version of *The Song of Enlightenment*, under study, has been shown through initial experiments to have been printed using movable metal type 786 years ago in 1239. If this is indeed the case, it is possible that the liquid printing substance (ink) containing trace amounts of metal could have chemically interacted with the metal type, transferring and fixing these metal traces onto the characters printed onto Korean paper (*Hanji*). Alternatively, metal elements on the surface of the metal type may have been transferred to the paper through direct

contact, as the back of the paper was rubbed while in contact with the movable metal type. (This is the traditional method of printing on paper in Korea.) If metal elements remain in the printed areas—such as the characters and border lines—there is a strong possibility that trace amounts of these metals can be detected through XRF measurements.

Quantitative scientific data on the metal elements present in the book will also provide insights into the likely printing method used. By analyzing the specific types of metal components detected, it may be possible to gather additional information about the type of metal used in casting the metal type. Figure 4 illustrates the first page (the front side of the folded first leaf) of the *Gongin* version of *The Song of Enlightenment*, being measured using a portable XRF device.

XRF measurements and element analysis were performed on all printed pages of the *Gongin* version of *The Song of Enlightenment*. To examine the differences in chemical element composition between the blank paper and the printed areas (characters and border lines), careful attention was given to selecting the measurement locations. A series of vertical line scans were conducted, starting from the top of the book and moving to the bottom along the printed characters. The measurement area was set to approximately 3 mm in diameter, the smallest area measurable by the portable XRF device used in this study. To enhance the signal-to-noise ratio (S/N), two types of X-ray beams were employed: Beam 1 (0.5 – 10 keV) and Beam 2 (0.5 – 40 keV). The analysis focused on the detection and comparison of 16 elements (Mg, Al, Si, P, S, K, Ca, Ti, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Sb) based on the XRF spectra.

The Chinese characters printed in the *Gongin* version of *The Song of Enlightenment* are generally square-shaped, measuring approximately 10 – 11 mm in both width and height, with stroke thickness ranging from 0.5 to 1.0 mm. Given that the XRF measurement area is 3 mm in diameter, no individual printed area exceeds this size. As a result, each XRF measurement captures signals from both a portion of the blank paper and the printed characters. In some instances, the inked area covered about 30% of the XRF measurement area. Consequently, the data from the printed character areas represents an average of the inked and blank paper signals. This means the difference between the printed and blank areas is somewhat underestimated. While the 3 mm diameter spatial resolution limits the ability to create detailed element maps using the portable XRF device, this experiment is nonetheless significant. It serves as a pivotal attempt to determine whether movable metal type was used in the printing of a book that has been a subject of scholarly debate for over fifty years, all while utilizing a widely accessible portable XRF measuring device.

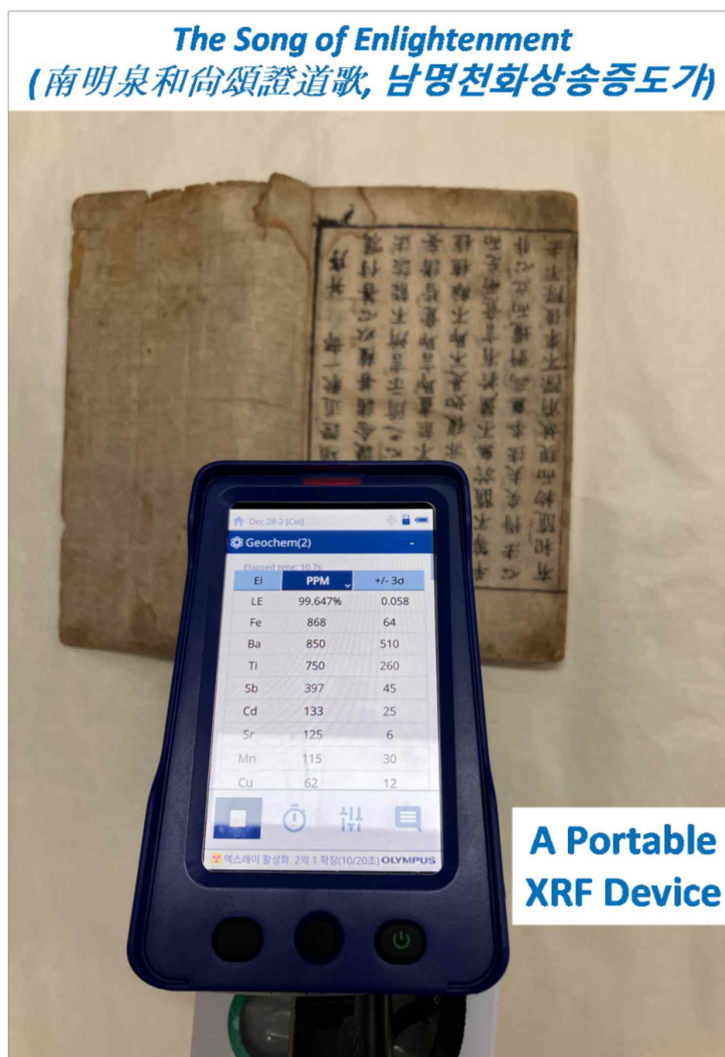


Figure 4. X-ray florescence measurement on the front side of the folded first leaf of *The Song of Enlightenment* (南明泉和尚頌證道歌). The book is upside down in the photograph. On the left side is the inside of cover page and on the right is the front side of the folded first leaf of the book. The XRF nose was directly placed on the desired spot to achieve the highest element sensitivity and measurement repeatability.

3. Results

3.1 Preliminary XRF Experiment using Hanji (Korean Paper) and Aluminum Foil

The *Hanji* paper, employed in the printing of the *Gongin* version of *The Song of Enlightenment*, is characterized by an exceptionally thin structure. Measurements of the individual sheets, prior to folding, reveal an average thickness of 87.3 μm . In the context of conducting X-ray fluorescence (XRF) analysis to assess the composition of the paper, it becomes essential to examine the extent to which X-rays penetrate the *Hanji* paper. Furthermore, the influence of the underlying stack of paper on the accuracy of the measurement warrants thorough

investigation. Understanding these factors is crucial for ensuring precise analytical results when using XRF for material composition analysis.

To evaluate the penetration depth of X-rays, eleven arrays consisting of aluminum foil and Hanji paper stacks were prepared. The aluminum foil, with an average thickness of 18.4 μm , and the *Hanji* paper, with an average thickness of 108.2 μm , were utilized. X-ray fluorescence (XRF) spectra were obtained from several configurations: seven sheets of aluminum foil placed atop eight sheets of *Hanji* paper, one sheet of aluminum foil placed on top of the *Hanji* paper stack, one sheet of aluminum foil positioned at different locations within the eight-sheet *Hanji* paper stack, and eight sheets of *Hanji* paper without any aluminum foil, as illustrated at the bottom of Figure 5. The presence of aluminum foil was detected through a stack of up to three sheets of *Hanji* paper. This observation confirms that the X-ray penetration depth is approximately three to four sheets of *Hanji* paper, corresponding to a range of 87.3 mm (261.9 – 349.2 mm).

Figure 6 presents the elemental composition of thirteen elements (Mg, Al, Si, P, S, K, Ca, Ti, Mn, Fe, Ni, Cu, and Zn) across the eleven arrays of Al foil(s) and *Hanji* paper stacks employing the portable XRF device. Significant concentrations of Si and Ca, exceeding 1,000 ppm (0.1%), were observed in all arrays, including those with Al foil(s) on top and those consisting solely of *Hanji* paper. As the number of *Hanji* paper layers atop a single sheet of Al foil increases, the Ca content also progressively rises. This trend serves as an indication of X-ray penetration depth, reaching up to eight stacks of *Hanji* paper sheets, with a total thickness of 698.4 μm . Notably, *Hanji* paper contains more than 10 ppm (0.001%) of Si, P, S, Ca, Ti, Mn, Fe, Ni, and Zn, as evidenced by the last bar in each element category, corresponding to the sample structure ID of 11. These findings demonstrate that *Hanji* paper contains low, but detectable levels of metallic elements.

The *Hanji* paper manufacturing process involves numerous factors that influence its quality and composition, ranging from the collection of raw materials, such as mulberry bark, to the use of special additives unique to each individual workshop, as well as the extensive use of local water. Raw materials like mulberry bark and the substantial quantities of local water are inherently subject to regional variations in trace elements. Soil and water quality differ across regions, and even when fibers for *Hanji* production are derived from the same species of mulberry tree, slight variations in properties and chemical compositions can occur depending on the geographical location. Additionally, groundwater and river water contain minerals of varying types and concentrations, further contributing to these regional differences.

Establishing a baseline for each chemical element in the blank paper of the book of interest is essential for accurate interpretation of XRF measurement results. If the *Hanji* paper originates from different craftsmen, workshops, locations, or eras, the baseline of each element in uncontaminated *Hanji* paper must be experimentally determined before interpreting XRF measurement results.

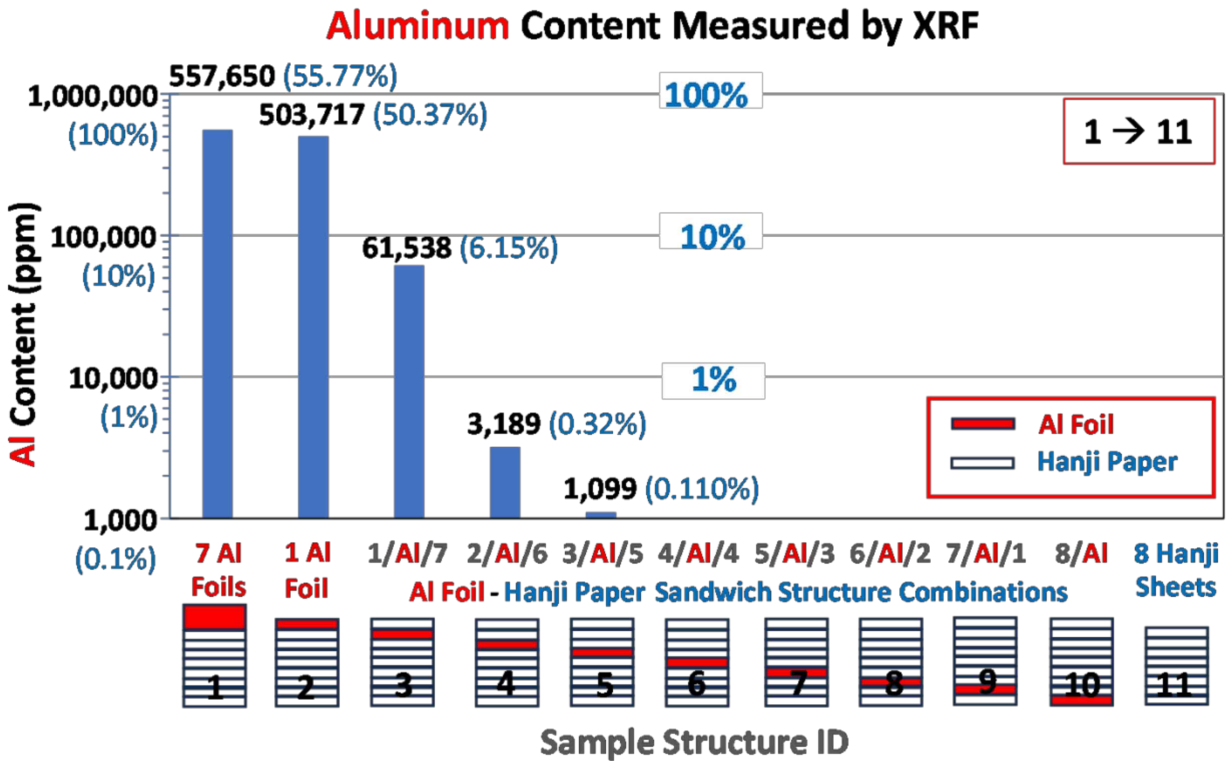


Figure 5. Al content measured from eleven arrays of Al foil and *Hanji* paper stacks using a portable XRF device. Al signal was detected from three-layer *Hanji* paper stack on one Al foil.

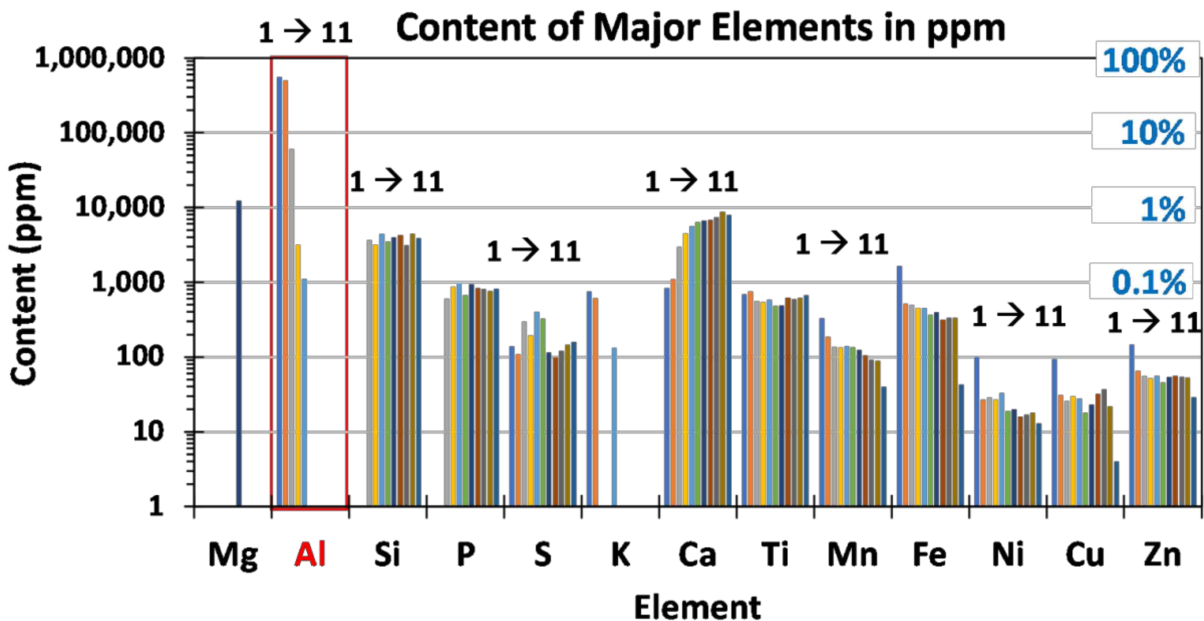


Figure 6. Measurement of thirteen elements from eleven arrays of Al foil and *Hanji* paper stacks using a portable XRF device. Significant amounts of Si and Ca, exceeding 1,000 ppm (0.1%),

were measured in all arrays. As the number of *Hanji* layers increases, the Ca content gradually increases.

3.2 XRF Measurement of The Song of Enlightenment

Figure 7 presents the first page (front side of the folded first leaf) of The Song of Enlightenment. Ten XRF measurement locations are indicated as red open circles, with the diameter of each circle approximating the area of the XRF measurement. Measurement sites 2, 3, 8, and 10, which display a dark brown color, show signs of water damage accumulated over nearly 800 years since the book's printing in 1239. There may also be an effect from organic contamination resulting from contact with readers' hands, primarily fingers, over multiple generations. Measurement sites 1, 5, and 10 are highlighted with square borders in different colors, representing the paper area without ink or water damage, the inked character, and the severely water-damaged areas, respectively. These sites correspond to the XRF spectra discussed in the following paragraph. In this study, the ink refers to the traditional Korean ink (*meog*, 墨), made from oil soot, and animal glue. The bar graphs in Figure 6 display the content of three elements (S, Fe, and Cu) measured by XRF at the ten sites. A summary of the XRF analysis for 23 elements across the ten sites is provided in Table 1, with element content reported in ppm.

In the beginning of XRF experiment, we have measured chemical elements of 27 different coins from various countries (Korea, Japan, Taiwan, Singapore and USA) and 20 modern Hanji papers to establish the baseline. We also rubbed some coins to *Hanji* papers to see possible contamination to the Hanji papers. We have detected noticeable amounts of metal contamination to the Hanji paper after rubbing metallic coins. To eliminate the possible metallic contamination including Fe and Cu from careless experimental procedures, Al foil experiment with Hanji paper is designed as seen in Figure 6 (Measurement IDs: 1 to 11). XRF measurement sensitivity of metallic element in Al foil covered with seven and one sheet of thin Hanji paper (Measurement IDs: 1 and 2) significantly. Almost one order of magnitude reduction of Mn, Fe, Ni, Cu and Zn contents were measured from the Hanji paper covered Al foil (Measurement IDs: 3 to 11). It is a proof of XRF sensitivity in depth direction. The element of interest at the XRF nose flush gives the highest element sensitivity. We also have measured XRF signals from 20 kinds of modern *Hanji* papers and 22 ancient books from the 13th to the 19th centuries in Korea.

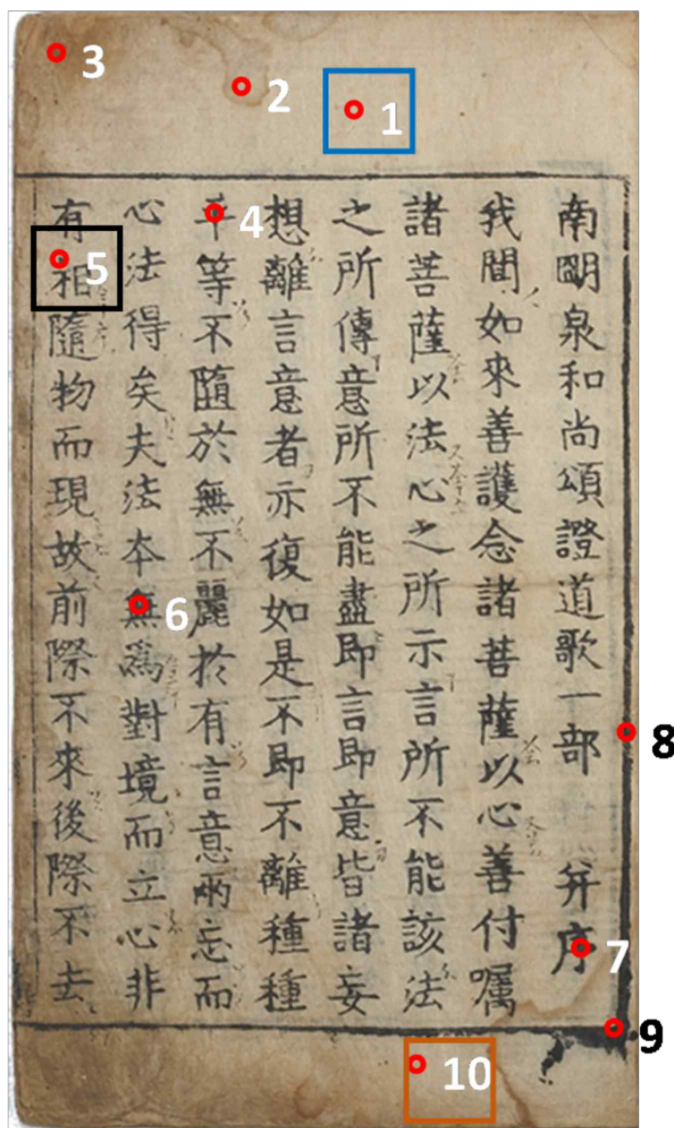
The Song of Enlightenment book is the original and no sign of repair was observed. We have measured XRF spectra at more than 500 points in the area of interest next to the inked area and one paper area without ink from all 88 pages. In Figure 7, the red horizontal line in each bar graph serves as a reference, facilitating the comparison between measurements from the relatively clean paper area, with no ink, and other measurement locations. All three elements (S, Fe, and Cu) measured from the clean paper area (measurement site 1) exhibited the minimum content across all measurement sites. Measurement sites 2 and 3, which show slight water damage, displayed a small increase in the content of all three elements (S, Fe, and Cu). Sites 4 through 7, where printed characters are located, showed a notable increase in S and Fe contents, with a relatively smaller increase in Cu content. Sites 8 and 9, on the printed border line, also exhibited a significant increase in S and Fe contents, with no or minimal increase in Cu content. Site 9 is affected by both ink from the printed border line and lighter water damage. The heavily water-damaged blank paper area in the bottom right corner (site 10) exhibited the highest content

for all three monitored elements, S, Fe, and Cu. Measurement sites (3, 4, 9, and 10), which display water damage, are marked with yellow arrows pointed downward for easier comparison.

The red horizontal line in each bar graph in Figure 7 is the average concentration of elements measured from the relatively clean paper area. It can be used as a reference to compare individual measurement results. It can be served as an alternate to error bars.

The Song of Enlightenment (南明泉和尚頌證道歌, 남명천화상송증도가)

Front Side of First Leaf



XRF Measurement Results

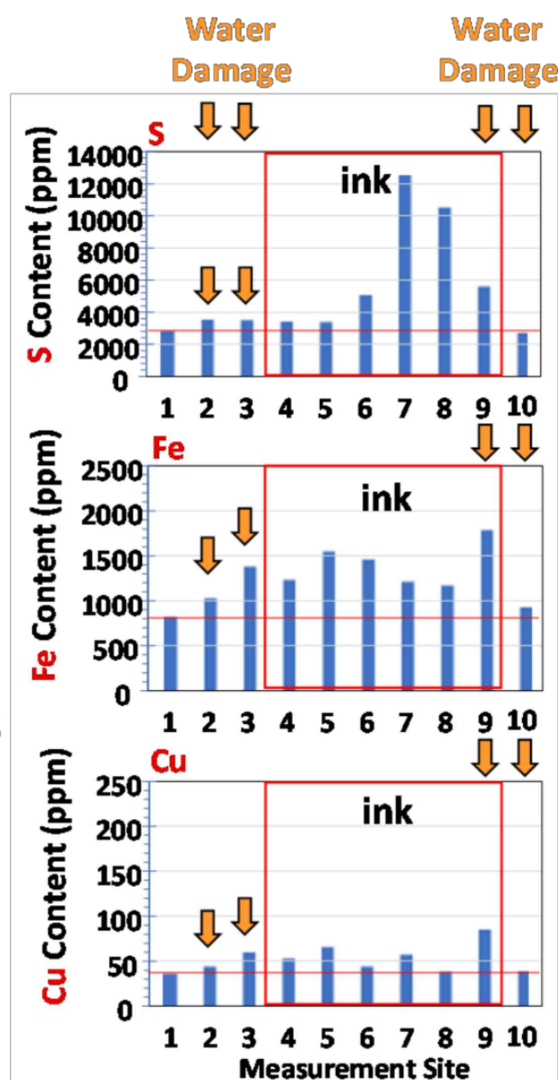


Figure 7. XRF measurement locations on the front side of the first leaf of *The Song of Enlightenment*. Measurement sites 2, 3, 8, and 10, highlighted in dark brown, exhibit signs of

water damage. The bar graphs present the elemental content of sulfur (S), iron (Fe), and copper (Cu) at the ten measurement sites.

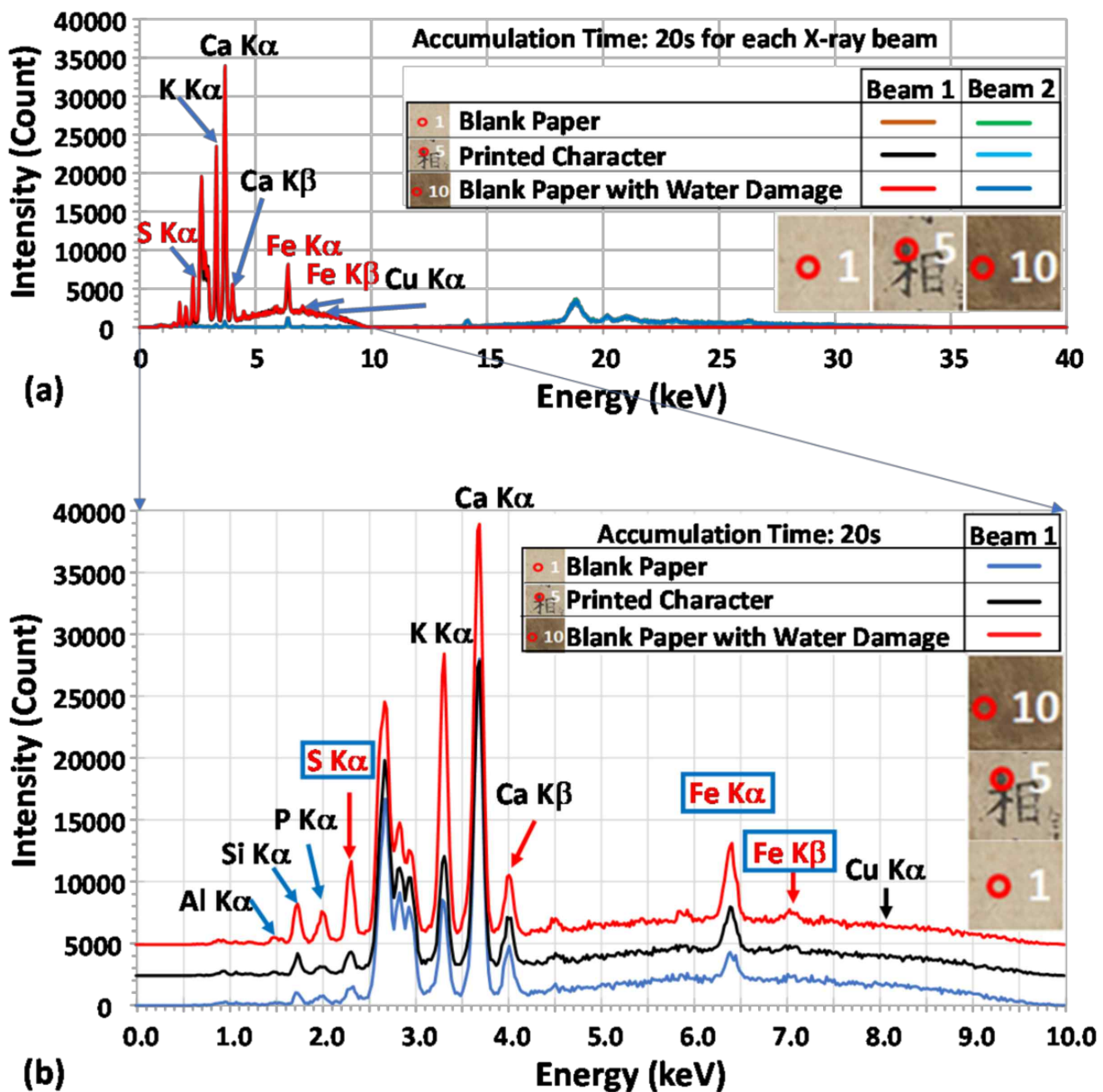


Figure 8. (a) Dual-beam XRF spectra (0.5 – 40 keV) and (b) low-energy XRF spectra (0.5 – 10 keV) from measurement sites 1, 5, and 10, with corresponding peak labels.

XRF spectra from measurement sites 1, 5, and 10, within the range of 0.5 – 40 keV, as well as detailed spectra in the range of 0.5 – 10 keV, are presented in Figure 8, with corresponding peak labels. Changes in the areas under the Al, Si, P, S, K, Ca, and Fe peaks are readily apparent. The quantity of each element is determined by the area under the corresponding

peaks, and the content is calculated by the area ratio of individual peaks to the total area. In cases where peaks are located very close together and the baseline of each peak is unclear, the height and position of individual peaks are determined through mathematical deconvolution. This process is fully automated by the software provided by the portable XRF device manufacturer. The raw XRF spectra and element analysis results can be exported in spreadsheet format for review, verification, and further customized analysis. The element analysis results, when compared with the corresponding source XRF spectra, confirm the reliability of the results. Therefore, the XRF measurement results for *The Song of Enlightenment* are both verifiable and robust, with a clear, documented process that ensures consistency and transparency.

One additional factor to consider in the interpretation of XRF measurement data is the penetration depth of X-rays ($\sim 300\text{ }\mu\text{m}$) in *Hanji* paper. The XRF measurement results represent a weighted average of 3 to 4 sheets, each $87.3\text{ }\mu\text{m}$ thick, below the measuring surface. The presence of printed characters or water damage beneath the measuring surface may also influence the measurement results. To accurately interpret XRF data from thin sheets of paper, particularly those thinner than the X-ray penetration depth, visual inspection of both the backside and any stacked papers (or other materials) beneath the measurement surface is essential.

Table 1. Summary of elemental analysis results from XRF measurements at ten sites, presented in ppm.

[illegible]

Pb	10	13	16	14	24	7	25	25	23	22
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Note: LOD stands for limit of detection

We believe that the repeated water damage and handling originated contamination at the bottom of book for about 800 years is the reason for the highest content for the three elements of S, Fe and Cu. According to the manufacturer of the XRF Analyzer, the interference-free Limit of Detection (LOD) for copper (Cu) typically ranges from 10 ppm to 50 ppm depending on the sample matrix and testing time. LODs for S and Fe are known to be fractions of the actual measured values S: 100 ppm – 500 ppm (0.01% - 0.05%) and Fe: 5 ppm – 20 ppm, respectively. Thus, the measurement values are considered to be reliable. The XRF spectra are analyzed automatically by deconvolution and curve fitting. The concentration of elements of interest is given as spread sheet. The Cu concentration measurement values for measurement sites 1 – 10 were ranging from 38 to 210 ppm as seen in Table 1. The measured S and Fe concentration were at least one order of magnitude higher all 10 sites. We believe that all the measurement is valid.

3.3 XRF Line Scan of *The Song of Enlightenment*

We have measured approximately ~600 points (= ~50 points/page x 12 pages) as line scans. The line can measurement was done for the front side of first 6 and last 6 folded leaves. We only reported the one of many sets of measurement to not to provide biased opinion. That was the reason why we selected XRF point measurement and line scan result on the printed page 1 in Figure 9.

XRF line scan measurements were conducted along the characters printed on the first line of the first page (front side of the folded first leaf) of the subject book, as shown in Figure 9. To avoid bias-related arguments over the measurement results, 44-points line scan spaced 5 mm apart, from top to bottom of the first printed page was summarized. A total of 44 measurement points, spaced 5 mm apart, were recorded along a 21.5 cm section, extending from the top to the bottom of the page, which measures 24.1 cm in height. The stitched line scan measurement field image, with measurement locations highlighted by small red circles, appears above the first line of the first page, where the XRF line scan was performed. The long red arrow indicates the direction of the XRF line scan. A bar graph summarizes the Fe content (in ppm) at the 44 measurement sites. The position of each bar in the graph corresponds to its approximate location along the red arrow.

The bar graph illustrates that all printed areas and the water-damaged blank paper at the bottom of the page (right end) exhibit Fe content 50 - 100% higher (1,000 - 1,440 ppm) compared to the baseline. The baseline Fe content, indicated by a red horizontal line at ~720 ppm was measured from three sites in the blank paper with no visible water damage at the top of the page (left side). Measurement site 4, which lacks printed characters or border lines on the front side, but is in close proximity to the printed border on the backside of the folded paper, shows higher Fe content than the three other sites (1 – 3). This indicates that the elevated iron (Fe) content at site 4 is likely influenced by the printed characters on the backside of the paper, similar to how the printed areas on the front side affect the Fe content. The presence of printing near the site on the backside most likely contributes to the higher Fe levels observed at this here. Measurement sites 31 – 33, do not contain printed characters on the front side, however, display

Fe content higher than the baseline. Notably, these sites, 31 – 33 contain printed characters on the backside of the paper and are stacked with papers that have printed characters beneath them.

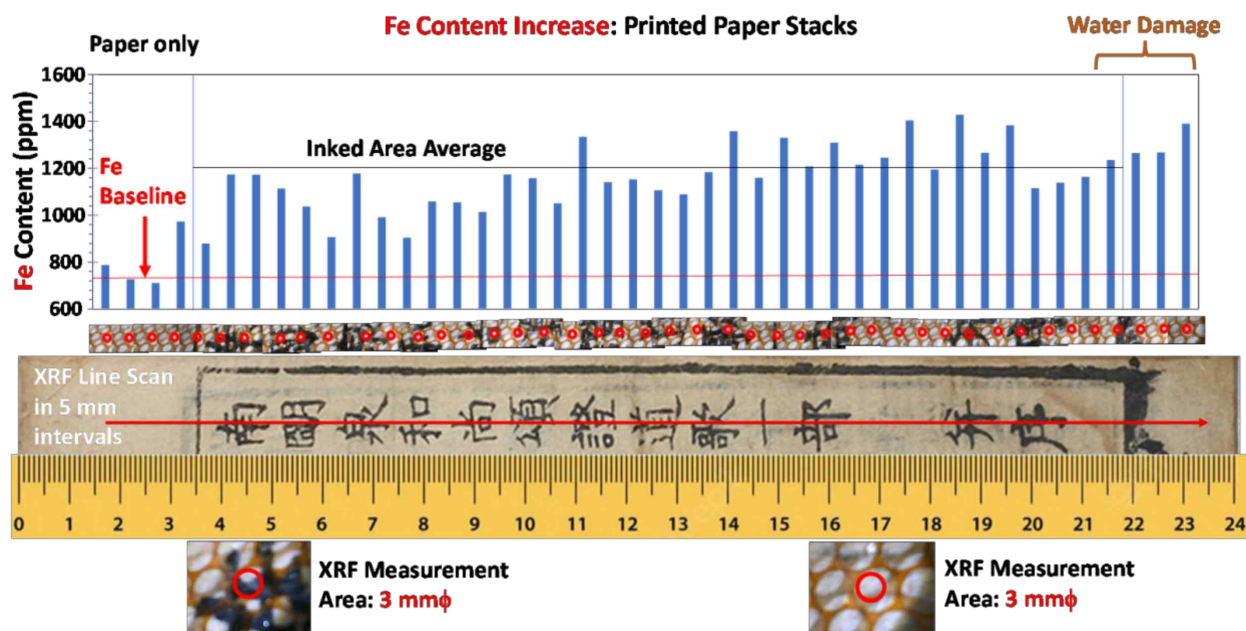


Figure 9. Summary of XRF line scan results for Fe content along the first line of printed characters. XRF spectra were measured at 44 points, spaced 5 mm apart, from top to bottom. The stitched measurement field image displays the line scan measurement sites, with each measurement location highlighted by small red circles. Two XRF measurement site images of 3 mm diameter are shown at the bottom.

Based on the significant increase in Fe content at XRF measurement sites from 4 to 42 within and near the printed area, the *Gongin* version of *The Song of Enlightenment*, which has been under controversy for more than half a century, can be concluded to be a metal type printed book from 1239, as stated in the colophon. Physics Magazine, based in Vancouver, reported an article titled “*The History of the Printing Press under an X-Ray Microscope*,” which highlighted a successful case of synchrotron imaging of historical and homemade prints at SRL, exploring possible connections between early printing methods in Korea and Europe (Berkowitz 2023). XRF element imaging using advanced equipment and/or synchrotron radiation facilities, such as SRL at Stanford University, USA (Edwards et al. 2022; SLAC 2025; Wagner et al. 2023; Berkowitz 2023), and ESRL in Grenoble, France (ESRF 2025), is highly recommended for further validation. Discussions regarding additional experiments abroad have taken place with the collector, as well as with Korea Heritage Service, and a positive response has been received from both. Efforts to seek a second verification from potential global collaborators are currently underway.

Currently housed at the French National Library in Paris, the ancient Korean book *Jikji*, printed in Korea in 1377, is and recognized by both UNESCO and the Guinness World Records as the oldest extant movable metal type book. The first documented use of movable metal type

for printing books took place in 1234, when it was used to print Confucian texts during the Goryeo dynasty in Korea. [9, 10, 26]. During the 143 years between these two important events, numerous books were printed with movable metal type in Korea, and it is reasonable to assume that extant examples of these books exist in Korea. One author recently discovered a movable metal type printed book from the 1360s, which was reported in a Korean academic journal (Yoo 2024b). This discovery predates *Jikji* by approximately 10 years. In January 2025, the American Printing History Association (APHA) updated the History of Printing Timeline to include this newly discovered book as the world's second-oldest movable metal-type printed book ([Korea Heritage Service 2023](#)). In 2019, two additional movable metal-type printed books were identified in Korea: one from the end of the Goryeo dynasty (918 – 1392) and the other from the early Joseon dynasty (1392). The one book titled *Singanyupyeon Yeokgeosamjang Munseondaechaek* (신간유편역거삼장문선대책 권5~6, 新刊類編歷舉三場文選對策 卷五~六) has been designated as a Korean Treasure [46]. Newly discovered books that have been identified as being printed with movable metal type have been the subject of ongoing controversy for the last 50 years. Judgments based on the expertise and insights of historians and bibliographers are significant and this case provides an opportunity for verification through scientific methods by these very people. There is a strong possibility that, in the future, even older books printed with metal type will be discovered in Korea, predating *Jikji* or *Sangjeong Gogyeum Yemun*. The latter is currently recognized as the earliest known movable metal-type printed book in 1234 ([Guinness World Records 2025a](#); [Chon 2025](#); [Guinness World Records 2025b](#); [Sohn 1959](#); [Sohn 1971](#); [American Printing History Association \(APHA\) 2023](#)).

4. Conclusions

The printing technique of an old book, with a colophon stating it was printed with movable metal type in 1239 during the Goryeo (高麗) dynasty of Korea, was investigated. The book is one of many versions of *The Song of Enlightenment* (南明泉和尚頌證道歌). This particular version, known as the *Gongin version* (空印本), has been classified as a duplicated woodblock printed book and was designated as a Korean Treasure in 2012. Although the colophon of the book asserts that it was printed with movable metal type in 1239, some researchers continue to interpret its contents as having been printed with duplicated woodblocks. This more than half-century-long controversy must be resolved with conclusive evidence.

To address this question, a series of experiments were conducted to detect any metal elements remaining in the old book using a portable X-ray fluorescence (XRF) device. Metal elements were detected from and near the printed characters. Notably, iron (Fe) content was found to be up to twice as high (~1,530 ppm) in characters printed with traditional Korean ink (meog, 墨) compared to plain paper (~800 ppm). Sulfur (S) content varied significantly, ranging from 2,300 ppm to 13,000 ppm across different measurement sites. Copper (Cu) and antimony (Sb) contents varied between 20 ppm and 90 ppm, and 300 ± 50 ppm, respectively. The Fe and S content were consistently higher in the printed characters and their immediate surroundings across all pages of *The Song of Enlightenment*. Based on these findings, the old book under

investigation was most likely printed using movable metal type in 1239, as stated in the colophon. Similarly, the printing method of any old book can be effectively assessed using quantitative XRF analysis data from both the printed areas and the blank paper. A portable XRF device proves to be a valuable tool for both cultural heritage applications and for determining the printing techniques employed in historical books.

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Author contributions

Authors are listed in descending order by significance of contribution.

The corresponding author is WSY.

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Methodology: WSY, SYW;

Software: WSY;

Validation: WSY, HJK;

Formal Analysis: WSY, SYW;

Investigation: WSY, SYW;

Resources: WSY, HJK;

Data Curation: WSY;

Writing – Original Draft Preparation: WSY;

Writing – Review & Editing: HJK;

Visualization: WSY, SOW;

Supervision: HJK.

Competing Interests

The authors declare that they have no competing interests.

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