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Optimizing the Solvent-to-Coffee Ratio for Caffeine Extraction from Arabica Kintamani Coffee Beans using Ethyl Acetate: A Comprehensive Study

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ABSTRACT

This paper studies the best ratio of solvent-to-coffee for getting caffeine from Arabica Kintamani coffee beans. This study aims to determine the optimal ratio of Kintamani Arabica coffee beans to ethyl acetate solvent for extracting caffeine, with the goal of improving the efficiency and quality of the extraction process. Previous research has not explored the best ratio for this specific application. The methodology employed in this investigation consists of four main steps: preparing materials, extracting and measuring caffeine, and analyzing the results. The paper provides the experimental data and discusses the impact of different ratios on the caffeine content in Arabica Kintamani coffee beans. Statistical analysis is used to demonstrate significant differences between the ratios, and the Tukey test is employed to compare them. The findings of this study indicate that the best solvent-to-coffee ratio for maximizing caffeine in ethyl acetate extracts is 1:5, which results in a concentration of 1930.9 ppm. This ratio provides the optimal balance between caffeine yield and solvent usage, ensuring the highest possible caffeine content while minimizing the amount of solvent required..

Keywords: *Solvent-to-coffee ratio; Caffeine extraction; Arabica Kintamani coffee beans.*

1. INTRODUCTION

Coffee, a cherished beverage with a rich cultural heritage, is consumed by millions of individuals worldwide. The health benefits of coffee have been extensively studied, with one of the most significant findings being that regular consumption of coffee is associated with a reduced risk of developing type 2 diabetes [1]. The consumption of coffee has been linked to preserving or enhancing kidney function, as indicated by the eGFR (estimated glomerular filtration rate) measurement. Observational studies, including one Mendelian randomization study, have shown that higher coffee consumption is linked to improved kidney function, although the evidence remains inconclusive. Studies have also found that coffee consumption is inversely associated with diabetes, which may contribute to the protective effects on kidney function. Further investigation is necessary to establish the underlying mechanisms through which coffee consumption may protect kidney function, with a particular focus on the cellular level [1], [2], [3]. Additionally, research suggests that coffee consumption may provide a protective effect against the development of liver cancer. [4]. These findings highlight the potential health advantages associated with the consumption of coffee.

Although caffeine has been demonstrated to improve athletic performance, it is crucial to consider the potential disadvantages. Factors such as genetics, physical condition, and psychological makeup can influence how caffeine affects an individual's performance in sports and exercise [5]. The ingestion of caffeine can result in sleep disturbances and sensations of anxiety [5]. Excessive consumption of

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caffeine can lead to intoxication and potentially hazardous outcomes [6]. Caffeine consumption has been reported to impact hydration levels, with some studies suggesting it may have a dehydrating effect, while others indicate no significant impact. A study found that moderate levels of coffee consumption can provide similar hydrating qualities to water. However, excessive caffeine intake can increase the risk of dehydration [7]. Regular coffee drinkers can develop a tolerance against the diuretic effects of caffeine, which may contribute to the overall hydration balance. This tolerance is a result of the body's adaptation to the effects of caffeine, which can lead to a reduction in the adenosine-blocking-action of caffeine and changes in gene expression in the striatum. However, the evidence for tolerance to the stimulatory effects of caffeine in humans is inconclusive [8], [9]. It is noteworthy that caffeine and smoking frequently co-occur, and both have associations with mental health concerns [10]. Additionally, caffeine can negatively impact cardiac function, leading to arrhythmias [11].

In light of the aforementioned paragraph, it is crucial to investigate a decaffeination process to address the various drawbacks associated with caffeine. Previous research has been conducted on caffeine extraction from coffee beans using solvents, with varying types of coffee beans, solvents, and optimum solvent-to-coffee ratios. One study specifically focused on the roasting process of coffee beans and analyzed the chemical composition of coffee bean extracts using nuclear magnetic resonance (NMR) [12] . Nevertheless, this study did not specifically focus on caffeine extraction using solvents. Another study investigated the decaffeination process of green coffee bean extract [13]. Although the study mentioned a reduction in caffeine content during the roasting and decaffeination processes, it did not offer specific information about the solvent employed or the solvent-to-coffee ratio used in the extraction of caffeine from whole coffee beans using supercritical carbon dioxide [14]. The researchers investigated the decaffeination rates as a function of CO2 flow rate, temperature, and pressure. However, they did not specify the type of coffee beans or the optimum solvent-to-coffee ratio used in their study. They compared nine common coffee extraction methods and found that different coffee beans contained varying amounts of caffeine per cup of coffee [15]. The researchers did not explicitly state the type of solvent used or the solvent-to-coffee ratio. They mentioned that solvents such as chloroform, dichloromethane, ethyl acetate, and supercritical carbon dioxide can be employed for caffeine extraction from coffee beans [16].. Although the study mentioned the use of ethyl lactate, water, and ethyl lactate and water mixtures for the pressurized liquid extraction of caffeine and catechins from green tea leaves, it did not provide specific details about the optimum solvent-to-coffee ratio used in this process [17]. It was reported that a caffeine/catechin recovery ratio of 2.0 was achieved, but the specific coffee beans or the solvent-to-coffee ratio was not mentioned. Supercritical carbon dioxide is recognized as an environmentally friendly solvent for caffeine extraction, but the study did not provide information about the specific solvent-to-coffee ratio used in this process [18]. Supercritical CO2 decaffeination is commonly characterized as solvent-free [19]. Although the study mentions the use of supercritical CO2 for caffeine extraction, it does not offer specific details about the solvent-to-coffee ratio used in this process [20]. The study did not provide information about the specific solvent-to-coffee ratio used in the caffeine extraction process. utilized reversed phase HPLC to quantify caffeine in coffee infusions, but did not specify the solvent or solvent-to-coffee ratio used. A natural deep eutectic solvent (NADES) called choline chloride-sorbitol was employed to extract caffeine and chlorogenic acid from green coffee beans [21]. he researchers aimed to establish the optimal extraction method and tested three different preparations of the NADES. However, they did not specify the type of coffee beans or the solvent-to-coffee ratio used in the study. In a review article, two primary methods for producing decaffeinated coffee using solvents were discussed: direct solvent extraction of the beans and water extraction of the beans followed by solvent extraction of the caffeine from the water extract. The review did not provide specific details about the type of coffee beans or the solvent-to-coffee ratio used in these processes [22].

Previously, studies on caffeine extraction from coffee beans using solvents have offered valuable knowledge on the decaffeination process. However, there is a requirement for additional research to determine the most effective and efficient method for extracting caffeine from coffee beans using solvents. The specific type of coffee beans, solvent, and optimum solvent-to-coffee ratio are yet to be fully understood. The solvent-to-coffee ratio plays a crucial role in the efficiency, quality, and cost of caffeine extraction processes. A higher ratio can result in a greater caffeine yield, but also increased solvent consumption and waste generation. On the other hand, a lower ratio may lead to a lower caffeine

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yield, but also lower solvent consumption and waste generation. Consequently, it is essential to optimize the solvent-to-coffee ratio for caffeine extraction using ethyl acetate to achieve the best balance between caffeine yield and solvent consumption.

This investigation aims to explore the impact of varying solvent-to-coffee ratios on the caffeine content derived from Arabica coffee beans using ethyl acetate as the solvent. The study's objective is to establish the optimal ratio that maximizes caffeine content, thereby improving the efficiency and quality of caffeine extraction processes. This research intends to provide valuable insights into the extraction process, which in turn can facilitate the production of caffeine-rich extracts for various applications.

2. RESEARCH METHODOLOGY

The methodology in this research consists of four primary stages, as following: materials preparation, caffeine extraction and quantification, and result statistical analysis.

2.1 Preparation

In the early stages of material preparation, Arabica Kintamani coffee beans were acquired from a supplier and kept in an airtight container at room temperature to ensure their quality. A technical-grade ethyl acetate for the research. Additionally, a range of essential tools and materials were required for the study, such as a Soxhlet apparatus, a rotary evaporator, a digital balance, a volumetric flask, a pipette, a UV spectrophotometry, and distilled water. Each of these items played a crucial role in facilitating the smooth and effective execution of the experiments..

2.2 Caffeine Extraction

To obtain caffeine from coffee beans, the beans were meticulously crushed using a coffee grinder until they reached the smallest possible particle size. The resulting powdered coffee was then sieved through a 60-mesh sieve to ensure uniformity. The exact weight of the strained coffee powder was measured using a digital scale and transferred into a thimble for further extraction procedures. To extract caffeine, a specific amount of ethyl acetate was measured using a measuring flask and carefully added to a round-bottom flask. The coffee powder, wrapped in filter paper, was then placed in the round-bottom flask along with the ethyl acetate. The flask was sealed tightly and placed in a water bath maintained at a consistent temperature of 50 degrees Celsius. The extraction process was allowed to continue for 2 hours without interruption. The extraction duration was influenced by the solvent-to-coffee ratio, which was investigated using five different ratios: 1:5, 1:7.5, 1:10, 1:12.5, and 1:15 (w/v). Each experiment was conducted as a single batch to ensure accuracy and consistent results.

2.3 Caffeine Quantification

The extraction process was carried out using ethyl acetate as the solvent, which effectively separated the caffeine from the other components of the sample. The resulting solution was then heated until all the caffeine had solidified, allowing for the weight of the extracted caffeine to be accurately measured using a digital balance. The concentration of caffeine in the ethyl acetate extract was calculated by dividing the weight of the extracted caffeine by the volume of ethyl acetate used during the extraction. To confirm the accuracy of the caffeine content in the ethyl acetate extracts, a standardized method utilizing UV spectrophotometry was employed. This involved dissolving predetermined quantities of pure caffeine in ethyl acetate and measuring their corresponding absorbance values to establish a calibration curve. The relationship between absorbance values and caffeine concentration was then used in linear regression to estimate the caffeine concentration in the ethyl acetate extracts.

2.4 Result Statistical Analysis

The analysis utilized statistical methods, including a one-way analysis of variance (ANOVA) to identify significant differences and a post-hoc Tukey test to pinpoint variations between individual ratios. The Real Statistics Resource Pack software (Version 8.7), developed by Charles Zaiontz as an add-in for Microsoft Excel 2019, was employed for the computations. In this assessment, any p-value below 0.05 was considered statistically significant.

3. ANALYSIS AND RESULT

The experimental findings are detailed in Table 1, which displays the caffeine concentration derived from Arabica coffee beans using various solvent-to-coffee ratios, as determined by UV Spectrometry. Figure 1 provides a visual representation of the average values and associated measurement errors for each set of solvent-to-coffee ratios.

Table 1. Concentration of Caffeine Extracted from Kintamani Arabica Coffee Beans Using Different Solvent-to-Coffee Ratios

Time (mins)	Concentration (ppm)				
	1:15	1:12.5	1:10	1:7.5	1:5
0	0.000	0.000	0.000	0.000	0.000
20	18.060	12.851	8.006	10.520	7.339
40	20.251	17.234	10.372	9.899	9.571
60	24.506	19.459	10.483	9.763	9.082
80	21.977	20.348	12.622	10.223	10.563
100	26.304	20.518	14.026	11.729	10.847
120	24.067	20.597	15.880	13.644	11.624

The study's results indicate that the amount of caffeine extracted from Kintamani Arabica coffee beans using ethyl acetate as the solvent is dependent on the ratio of solvent to coffee. As the ratio of solvent to coffee increases, the concentration of caffeine extracted also increases. This suggests that a higher quantity of solvent leads to a greater extraction of caffeine from the coffee beans.

The descriptive statistics presented in Table 2 offer a comprehensive analysis of the characteristics of each group, highlighting the variations and trends within each set. The histograms in Figure 1, which depict the variance and standard error for each group, provide a visual representation of the data, allowing for a more in-depth understanding of the distributions. The results of the ANOVA test, as displayed in Table 3, indicate a significant difference in caffeine content among the various solvent-to-coffee ratios ($F(4,08724) = 0.0092$, $p < 0.05$), suggesting that these ratios have a significant impact on the caffeine content of the coffee..

Table 4 displays the results of the post-hoc Tukey test, which includes pairwise comparisons and p-values. The test reveals that the 1:5 solvent-to-coffee ratio has a significantly higher caffeine content compared to the 1:15 and 1:12.5 ratios ($p < 0.05$ for all comparisons). Additionally, the 1:7.5 ratio shows a significantly higher caffeine content compared to the 1:15 ratio ($p < 0.05$ for all comparisons). These findings indicate that the 1:5 ratio has the highest caffeine content among all ratios, as it is significantly higher than two other groups of solvent-to-coffee ratios. It is noteworthy that there are no significant differences in caffeine content between any pair of groups with p-values of 0.05 or greater.

The research indicates that the proportion of solvent to coffee plays a crucial role in the effectiveness of caffeine extraction using ethyl acetate as the solvent. The most effective solvent-to-coffee ratio for maximizing caffeine content in ethyl acetate extracts is a ratio of 1:5, which results in a concentration of 1930.9 mg/mL. This ratio strikes an optimal balance between caffeine yield and solvent usage.

In summary, the study found that there was no significant difference in the extraction efficiency of caffeine when using ethyl acetate as a solvent at ratios of 1:10, 1:7.5, and 1:5. The p-value was greater than 0.05, indicating that the results were not statistically significant. The possible explanation for this finding is that coffee solids contain substances like oils, acids, sugars, and proteins that interact with ethyl acetate and reduce its ability to dissolve caffeine. As a result, the more solvent used may cause less caffeine to dissolve, leading to a reduced extraction efficiency.

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Table 2. Descriptive. Statistics for Different Groups

DESCRIPTION				Alpha	0.05
Group	*Count*	*Mean*	*Variance*	*SS*	*Std Err*
1:15	7	843.229	157626	945753	237.123
1:12.5	7	939.686	190180	1141082	237.123
1:10	7	1019.843	269550	1617301	237.123
1:7.5	7	1628.671	548973	3293840	237.123
1:5	7	1930.92	801634	4809805	237.123

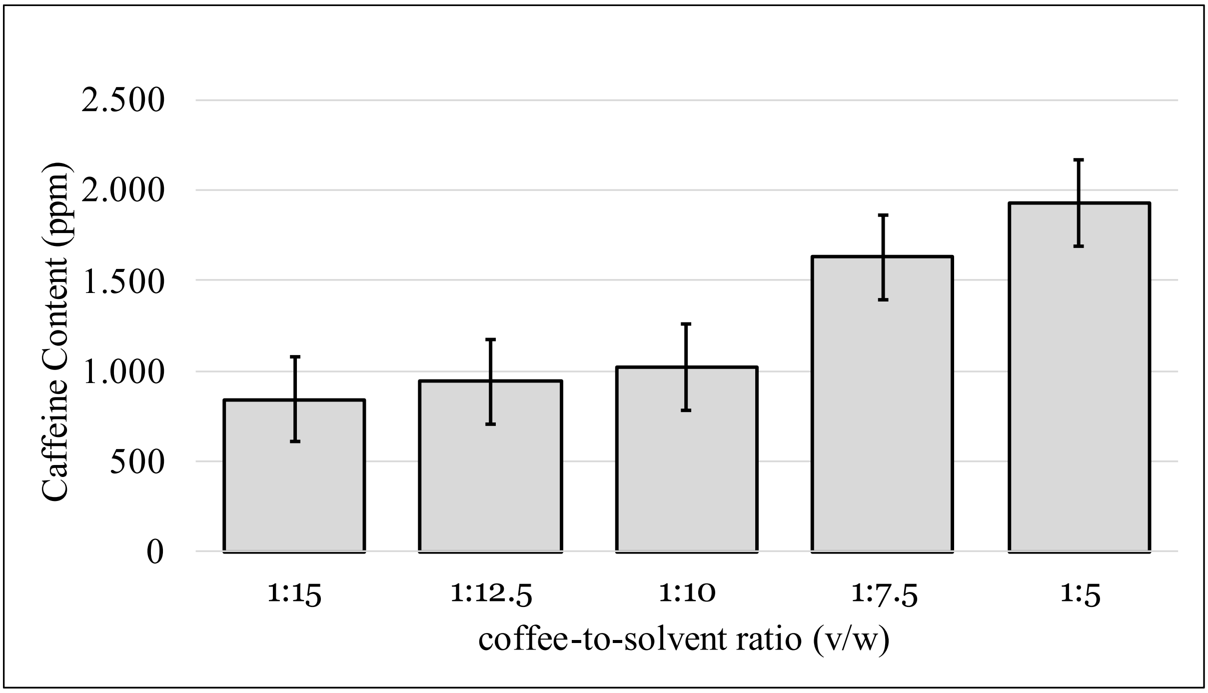

Figure 1. Mean Values and Standard Errors for Each Group of Solvent-to-Coffee Ratios

Table 3. ANOVA Results for Source of Variation

ANOVA

Sources	*SS*	*df*	*MS*	*F*	*P value*
Between Groups	5497348.9	4.0	1374337	5.063	0.003
Within Groups	814340	30.0	271446,7		
Total	13640749,5	34.0	401198.5		

Table 4. P-Values for Pairwise Comparisons in Variable Groups

p-value	1:15	1:12.5	1:10	1:7.5	1:5
1:15	-	0.998	0.984	**0.160**	**0.022**
1:12.5	0.998	-	0.999	0.266	**0.044**
1:10	0.984	0.999	-	0.384	0.075
1:7.5	**0.160**	0.266	0.384	-	0.894
1:5	**0.022**	**0.044**	0.075	0.894	-

4. CONCLUSION

The caffeine content extracted from Arabica Kintamani coffee beans using ethyl acetate can be influenced by the solvent-to-coffee ratio. As the ratio increases, the caffeine concentration also rises. Descriptive statistics and ANOVA tests have confirmed that there are significant differences among the ratios. Among the tested ratios, 1:5 has the highest caffeine content, followed by 1:7.5. The optimal ratio for maximum caffeine extraction is 1:5, which results in a caffeine concentration of 1930.9 mg/mL. There are no significant differences in caffeine content between ratios 1:10, 1:7.5, and 1:5..

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REFERENCES

[1] K. Nakajima, K. Hirose, M. Ebata, K. Morita, and H. Munakata, "Association between habitual coffee consumption and normal or increased estimated glomerular filtration rate in apparently healthy adults," *British Journal of Nutrition*, vol. 103, no. 2, pp. 149–152, 2010, doi: 10.1017/S0007114509991681.

[2] M. Kanbay *et al.*, "Effect of Coffee Consumption on Renal Outcome: A Systematic Review and Meta-Analysis of Clinical Studies," *Journal of Renal Nutrition*, vol. 31, no. 1. W.B. Saunders, pp. 5–20, Jan. 01, 2021. doi: 10.1053/j.jrn.2020.08.004.

[3] E. A. Hu, E. Selvin, M. E. Grams, L. M. Steffen, J. Coresh, and C. M. Rebholz, "Coffee Consumption and Incident Kidney Disease: Results From the Atherosclerosis Risk in Communities (ARIC) Study," *American Journal of Kidney Diseases*, vol. 72, no. 2, pp. 214–222, Aug. 2018, doi: 10.1053/j.ajkd.2018.01.030.

[4] T. Shimazu *et al.*, "Coffee consumption and the risk of primary liver cancer: Pooled analysis of two prospective studies in Japan," *International Journal of Cancer*, vol. 116, no. 1, pp. 150–154, 2005, doi: 10.1002/ijc.20989.

[5] N. S. Guest *et al.*, "International society of sports nutrition position stand: caffeine and exercise performance," *Journal of the International Society of Sports Nutrition*, vol. 18, no. 1, 2021, doi: 10.1186/s12970-020-00383-4.

[6] M. H. Alfaifi *et al.*, "Assessment of caffeine consumption behavior among Jazan University students in the south of Saudi Arabia: A cross-sectional study," *Medicine (United States)*, vol. 101, no. 51, 2022, doi: 10.1097/MD.0000000000031651.

[7] S. C. Killer, A. K. Blannin, and A. E. Jeukendrup, "No evidence of dehydration with moderate daily coffee intake: A counterbalanced cross-over study in a free-living population," *PLoS One*, vol. 9, no. 1, Jan. 2014, doi: 10.1371/journal.pone.0084154.

[8] J. Watson, I. Deary, and D. Kerr, "Central and peripheral effects of sustained caffeine use: tolerance is incomplete," 2002.

[9] "Caffeine and Coffee Tolerance." [Online]. Available: http://ahajournals.org

[10] L. Schellhas *et al.*, "Maternal and child genetic liability for smoking and caffeine consumption

and child mental health: an intergenerational genetic risk score analysis in the ALSPAC cohort," *Addiction*, vol. 116, no. 11, pp. 3153–3166, 2021, doi: 10.1111/add.15521.

[11] M. Porta *et al.*, "Single ryanodine receptor channel basis of caffeine's action on Ca 2+ sparks," *Biophysical Journal*, vol. 100, no. 4, pp. 931–938, 2011, doi: 10.1016/j.bpj.2011.01.017.

[12] F. Wei, K. Furihata, M. Koda, F. Hu, T. Miyakawa, and M. Tanokura, "Roasting process of coffee beans as studied by nuclear magnetic resonance: Time course of changes in composition," *Journal of Agricultural and Food Chemistry*, vol. 60, no. 4, pp. 1005–1012, 2012, doi: 10.1021/jf205315r.

[13] S. J. Song, S. Choi, and T. Park, "Decaffeinated green coffee bean extract attenuates diet-induced obesity and insulin resistance in mice," *Evidence-based Complementary and Alternative Medicine*, vol. 2014, 2014, doi: 10.1155/2014/718379.

[14] H. Peker, M. P. Srinivasan, J. M. Smith, and B. J. McCoy, "Caffeine extraction rates from coffee beans with supercritical carbon dioxide," *AIChE Journal*, vol. 38, no. 5, pp. 761–770, 1992, doi: 10.1002/aic.690380513.

[15] A. N. Gloess *et al.*, "Comparison of nine common coffee extraction methods: Instrumental and sensory analysis," *European Food Research and Technology*, vol. 236, no. 4, pp. 607–627, 2013, doi: 10.1007/s00217-013-1917-x.

[16] A. Belay, "Measurement of integrated absorption cross-section, oscillator strength and number density of caffeine in coffee beans by integrated absorption coefficient technique," *Food Chemistry*, vol. 121, no. 2, pp. 585–590, 2010, doi: 10.1016/j.foodchem.2009.12.052.

[17] D. V. Bermejo, J. A. Mendiola, E. Ibáñez, G. Reglero, and T. Fornari, "Pressurized liquid extraction of caffeine and catechins from green tea leaves using ethyl lactate, water and ethyl lactate + water mixtures," *Food and Bioproducts Processing*, vol. 96, pp. 106–112, 2015, doi: 10.1016/j.fbp.2015.07.008.

[18] Y. Saotome and M. Imai, "Supercritical carbon dioxide extraction of apigenin from parsley leaves pre-treated to maximize yield," *Food Science and Technology Research*, vol. 24, no. 1, pp. 63–73, 2018, doi: 10.3136/fstr.24.63.

[19] T. Welton, "Solvents and sustainable chemistry," *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 471, no. 2183, 2015, doi: 10.1098/rspa.2015.0502.

[20] A. B. A. De Azevedo, P. Mazzafera, R. S. Mohamed, S. A. B. Vieira De Melo, and T. G. Kieckbusch, "Extraction of caffeine, chlorogenic acids and lipids from green coffee beans using supercritical carbon dioxide and co-solvents," *Brazilian Journal of Chemical Engineering*, vol. 25, no. 3, pp. 543–552, 2008, doi: 10.1590/s0104-66322008000300012.

[21] E. Yuniarti, F. C. Saputri, and A. Mun'im, "Application of the natural deep eutectic solvent choline chloridesorbitol to extract chlorogenic acid and caffeine from green coffee beans (Coffea canephora)," *J Appl Pharm Sci*, vol. 9, no. 3, pp. 82–90, 2019, doi: 10.7324/JAPS.2019.90312.

[22] V. Kumar, M. Madhava Naidu, and G. A. Ravishankar, "Developments in coffee biotechnology - In vitro plant propagation and crop improvement," *Plant Cell Tissue Organ Cult*, vol. 87, no. 1, pp. 49–65, 2006, doi: 10.1007/s11240-006-9134-y.