# Multielement ICP-OES Analysis of Mineral Premixes Used to Fortify Foods

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**ABSTRACT**

A method for the multielement analysis of mineral premixes using inductively coupled plasma optical emission spectrometry (ICP-OES) was developed and validated. Due to the lack of certified premix samples, the validation was performed on five different purchased premixes.

A microwave-assisted acid hydrolysis was used to bring the elemental composition into solution.

A standardized dilution scheme permitted the determination of different ranges of elemental concentrations found in premixes. Calcium, chromium, copper, iron, magnesium, manganese, molybdenum, selenium, and zinc were determined simultaneously by ICP-OES. Robust repeatability and intermediate reproducibility were estimated and found to be sufficient for three out of five premixes. The same was true for accuracy which had been estimated by the determination of recovery from spiked premixes. Insufficient repeatability and accuracy for two pet food premixes was due to their heterogeneity. Using a larger sample size for these premixes significantly improved the repeatability and intermediate reproducibility for several elements.

The developed multielement method performed well on homogeneous mineral premixes, but high repeatability data are to be expected from pet food premixes due to their high heterogeneity.

## INTRODUCTION

The fortification of foods with vitamins and minerals is one of the most effective methods to improve health and prevent nutritional deficiencies. This method has helped to eliminate diseases such as goitre, rickets, and beriberi in many countries. Examples of early fortified foods are iodized salt in the 1920s, milk with vitamin D in the 1930s, and flour and bread enriched with vitamins B1, B2 and Fe in the 1940s. Nowadays, fortification allows the standardization of nutrient content in foods that show natural variable concentrations (1).

It is obvious that the concentrations of vitamins and minerals in premixes have to be exact in order to ensure the quality of the finished products. In the case of Se, this is not only a quality issue but also a safety concern because the concentration range between beneficial and adverse health effects is narrow (2).

Analytical methods for minerals in foodstuff range from colorimetric methods to inductively coupled plasma mass spectrometry (ICP-MS). However, only a few methods report on the analysis of fortified foods or food supplements. A rapid method for iron determination in such foods has been described recently using spectrophotometry (3). The selenium concentration in dietary supplements is mainly checked by ICP-MS or hydride generation atomic absorption spectroscopy (HG-AAS) (4,5). In fact, atomic absorption spectroscopy (AAS) is one of the most commonly used tools for mineral determination in foods or animal feed and some methods have been reported in the literature (6–9). The major drawback of AAS, however, is that not all important elements can be measured with one system and that analysis is not simultaneous. Inductively coupled plasma optical emission spectrometry (ICP-OES) features multielement capability, wide linear dynamic range, high analytical sensitivity, and high sample throughput. Multielement analysis by ICP-OES has become routine and accuracy is better or at least comparable to AAS (10). No ICP-OES multielement analysis with regard to the problem of mineral premixes has been found in a recent literature survey. However, the simultaneous determination of minerals in multivitamin and salinated water samples has been reported (11,12). These studies address similar problems such as high salt concentrations and a high concentration range of minerals which have to be taken into account by appropriate dilutions. Table I lists the concentration range encountered in the investigated premixes.

It was the aim of this study to develop and validate a method using ICP-OES for the determination of Ca, Cu, Cr, Fe, Mg, Mn, Mo, Se, and Zn in mineral premixes. Acid microwave digestion was used for sample preparation, a widely used method in food sample digestion (13-16). Several emission lines were tested for interferences and correction by internal standards was applied for all elements in order to compensate for adverse effects of high salt charges. Limits of quantitation, robust repeatability, intermediate reproducibility, and accuracy (recovery of minerals from spiked premixes) were estimated.

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**TABLE I**
**Expected Concentration Range of Minerals in Most Premixes Used for Food Fortification**

| | Ca | Cr | Cu | Fe | Mg | Mn | Mo | Se | Zn |
|---|---|---|---|---|---|---|---|---|---|
| Min ($mg \cdot kg^{-1}$) | 8000 | 20 | 50 | 500 | 25000 | 1 | 15 | 10 | 500 |
| Max ($mg \cdot kg^{-1}$) | 80000 | 1000 | 40000 | 100000 | 40000 | 90000 | 15000 | 7000 | 90000 |

## EXPERIMENTAL

### Instrumentation

A CEM (MDS 2000, max. 630 W at 100% of Power) microwave digestion system with glass volumetric flasks was used for sample preparation.

For the nine element measurements, a Varian Vista-Pro Axial ICP-OES (Varian, Mulgrave, Victoria, Australia) was used. A cyclonic spray chamber (thermostated at 15°C) coupled with a Micromist nebulizer (Glass Expansion, West Melbourne, Victoria, Australia) was used as the injection device. The operating conditions are shown in Table II.

A solution containing internal standards (strontium and indium) was mixed on-line with analytical solutions using a mixing manifold (2 ways in and 1 way out) linked between the peristaltic pump and the nebulizer.

### Reagents and Standard Solutions

High-purity water (18.2 MΩ) using a Milli-Q™ Plus system (Millipore, Bedford, MA, USA).

Nitric acid freshly sub-distilled from conc. nitric acid (analytical grade, 65% p.a.). Suprapur® hydrochloric acid (30%) (Merck, Darmstadt, Germany). Suprapur hydrogen peroxide (30% v/v) (Merck).

Nine element standards were prepared by dilution of a 1000-mg $L^{-1}$ single-element stock solution (Merck) in a 5% $HNO_3$ matrix.

**TABLE II**
**Axial ICP-OES Operating Conditions**

| | | |
|---|---|---|
| Power | 1300 W | |
| Plasma gas-flow | 18 L $min^{-1}$ | |
| Auxiliary flow | 2.25 L $min^{-1}$ | |
| Spray chamber | Cyclonic, internal volume = 100 mL | |
| | Argon flow 0.90 L $min^{-1}$ | |
| Nebulizer | Micromist, Micro-concentric nebulizer | |
| | 1 mL $min^{-1}$ | |
| Replicate read time | 5 s | |
| Replicates | 5 | |
| Main emission lines (nm) | Ca 422.673 | Cr 283.563 |
| | Cu 324.754 | Fe 259.940 |
| | Mg 279.553 | Mn 257.610 |
| | Mo 202.032 | Se 196.026 |
| | Zn 213.857 | |
| | In 303.936 (internal standard, 40 mg $L^{-1}$) | |
| | Sr 338.071 (internal standard, 10 mg $L^{-1}$) | |
| Alternatively studied emission lines (nm) | Cr 205.560 | Cr 267.716 |
| | Mo 204.598 | Mg 280.270 |
| | Mg 285.213 | |

Strontium was prepared by dilution of a 1000-mg $L^{-1}$ Sr stock solution (Fluka, Buchs, Switzerland)

Indium was prepared by dilution of a 1000-mg $L^{-1}$ In stock solution (Merck).

### Sample Preparation

*Microwave Digestion*

One g of sample was weighed into 100-mL glass volumetric flasks. Five mL of 65% $HNO_3$ was added and left to react for half an hour at ambient temperature; then the volumetric flask was heated on a hot plate (150°C) until the acid digestion started (yellow fume production). Then 5 mL of 30% $H_2O_2$ was slowly added to prevent a fast reaction. After predigesting the samples, the flasks were put into the microwave oven and an appropriate power program was used, depending on the number of samples to be digested (see Table III). The first step required 20 min heating time. After this step, the flasks were carefully taken out of the microwave oven and 5 mL 30% HCl was added. The flasks were reheated for 10 min using the same program. After removing the flasks from the oven, they were left standing to cool, then each flask was brought to volume with high purity water. A preparation blank was included in each series of digestions.

**TABLE III**
**Microwave System: Digestion Program**

| No. of Vessels | Power (%) |
|---|---|
| 5 | 23 |
| 6 | 27 |
| 7 | 31 |
| 8 | 35 |
| 9 | 39 |
| 10 | 43 |
| 11 | 47 |
| 12 | 51 |

### Dilutions

Each digested sample was analyzed undiluted, 20 times diluted, and 200 times diluted. The dilutions were produced using a 5% $HNO_3$ solution.

## RESULTS AND DISCUSSION

### Calibration and Linearity

Calibration was performed using six standards with 0.2, 0.5, 1.0, 1.5, 2.0, and 5.0 mg $L^{-1}$ concentrations for Ca, Cu, Fe, Mg, Mn, Mo, and Zn and 0.1, 0.2, 0.4, 0.6, 0.8, and 1.0 mg $L^{-1}$ for Se and Cr. On our apparatus, the calibration type was a linear fit for all elements, with the exception of Mg, which followed a quadratic calibration model. All calibration curves had a $R^2 > 0.99$.

### Ruggedness-Study of Interferences

Wavelengths were chosen according to their least probability of interferences with other elements. Recoveries were determined from low concentration spikes of analytes (0.1 mg $L^{-1}$) in matrices with a high concentration of the other eight elements (25 mg $L^{-1}$). Recoveries were calculated using the following equation:

$$\%\text{Recovery} = \frac{[\text{Analyte} + 8\ \text{Elements}]}{\text{Analyte}} \cdot 100\%$$

**[Analyte + 8 Elements]** = Concentration of analyte found in the sample with the analyte and the 8 other elements

**Analyte** = Concentration of analyte in the sample without other elements

The recoveries were found to be between 95–110% for all elements, except for Zn (see Table IV). Zinc was strongly influenced by signals from Cu 213.854 nm and Fe 213.859 nm (Figure 1a and Table V). This was only true when Zn was present in low concentration compared to the interfering element. Zinc was not influenced when Fe and Cu were present at the same concentration (2.5 mg $L^{-1}$) (see Figure 1b and Table V) and therefore the Zn line at 213.851 nm was chosen for our method. Two alternative wavelengths for Cr were tested and resulted in good recoveries; but in order to obtain maximal sensitivity, the Cr 283.563 nm was chosen.

### Limits of Quantitation

Microwave digests of premix matrices (free of analyte) were spiked with low concentrations of the nine elements. The concentration of the lowest standard was chosen for these spikes. Limits of quantitation were calculated using the following equation and are listed in Table VI:

$$\text{LOQ} = \frac{[\text{Spike}]}{(A_{spike} - A_{matrix})} \cdot SD_{matrix} \cdot 10$$

**[Spike]** = Spike concentration in mg $L^{-1}$

$\mathbf{A}_{spike}$ = Peak height measured for the spike in the digested matrix.

$\mathbf{A}_{matrix}$ = Peak height measured for digested matrix expected free of analyte. In general this value was negligible compared with the peak height of the spiked sample.

$\mathbf{SD}_{matrix}$ = Standard deviation of peak height for digested matrix expected free of analyte.

**TABLE IV**
**Recovery From Interference Study of Nine Elements**
**The underlined wavelengths were used for the final method; recoveries in bold were accepted. All measurements were carried out in duplicate.**

| Analyte and Wavelength (nm) | Concentration | Depending on Internal Std In | Sr |
|---|---|---|---|
| Ca 422.673 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$all | **113** | 116 |
| Cr 283.563 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | **106** | 110 |
| Cr 205.560 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 101 | 105 |
| Cr 267.716 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 97 | 101 |
| Cu 324.754 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$all | **101** | 105 |
| Fe 259.940 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | **98** | 102 |
| Mg 279.553 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | **109** | 113 |
| Mg 280.270 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 108 | 113 |
| Mg 285.213 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 110 | 114 |
| Mn 257.610 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | **101** | 98 |
| Mo 202.032 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 94 | **99** |
| Mo 204.598 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 98 | 103 |
| Se 196.026 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | **98** | 106 |
| Zn 213.857 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 179 | 170 |

### Repeatability and Intermediate Reproducibility

Five premixes (A–E) were used for repeatability and intermediate reproducibility studies (see Table VII). Samples A–C were standard trace element premixes and D–E were pet food premixes (see Table VII). Robust relative repeatability limits (r%) (at 95% confidence interval) were calculated from 14 replicates per premix measured by the same operator at the same day; and the results are given in Table VII. The robust intermediate reproducibility limits (iR%) were calculated from duplicate analyses on six different days (carried out in the same laboratory and by the same operator) and are listed in Table VIII.

**TABLE V**
**Recoveries From Interference Study with Zn**
**(The underlined wavelengths were used for the final method; recoveries in bold were accepted. All measurements were carried out in duplicates. See text for details.)**

| Analyte and Wavelength (nm) | Concentration | %Rec Depending on Internal Std | |
|---|---|---|---|
| | | In | Sr |
| Zn 213.857 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ all | 179 | 170 |
| Zn 213.857 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ Cu | 153 | 152 |
| Zn 213.857 | 0.1 mg $L^{-1}$ + 25 mg $L^{-1}$ Fe | 124 | 120 |
| Zn 213.857 | 2.5 mg $L^{-1}$+ 2.5 mg $L^{-1}$ Cu | 100 | **100** |
| Zn 213.857 | 2.5 mg $L^{-1}$ + 2.5 mg $L^{-1}$ Fe | 101 | **101** |

**TABLE VI**
**Limits of Quantitation Determined on a Digested Premix Matrix**
**(Concentrations are given in mg $kg^{-1}$)**

| Ca | Cr | Cu | Fe | Mg | Mn | Mo | Se | Zn |
|---|---|---|---|---|---|---|---|---|
| 1.0 | 1.0 | 1.0 | 2.0 | 0.1 | 0.1 | 1.5 | 20 | 0.5 |

The elemental composition of the premixes is given in the next paragraph (Accuracy, Table IX). Only those elements were evaluated that had been declared by the manufacturer of the premix. Others were not measured and their cells in Tables VII–X were left empty (–).

Tables VII and VIII show that the robust relative repeatabilities and intermediate reproducibilities were found to be < 5% and < 16%, respectively, for premixes A–C. Premixes D and E had high robust relative repeatability and intermediate reproducibility, and were reanalyzed using 2.5 g as the sample size. The digestion method was adapted using 10 mL of $HNO_3$ and 10 mL of $H_2O_2$ for the first step in the microwave digestion. Microwave conditions and the added volume of HCl remained the same as in the above-described sample preparation. The samples were diluted twice with water in order to obtain the same $HNO_3$ concentration as present in the standards. Further dilutions were carried out based on these twice-diluted samples. The high repeatability and reproducibility data decreased through this adaptation of the method.

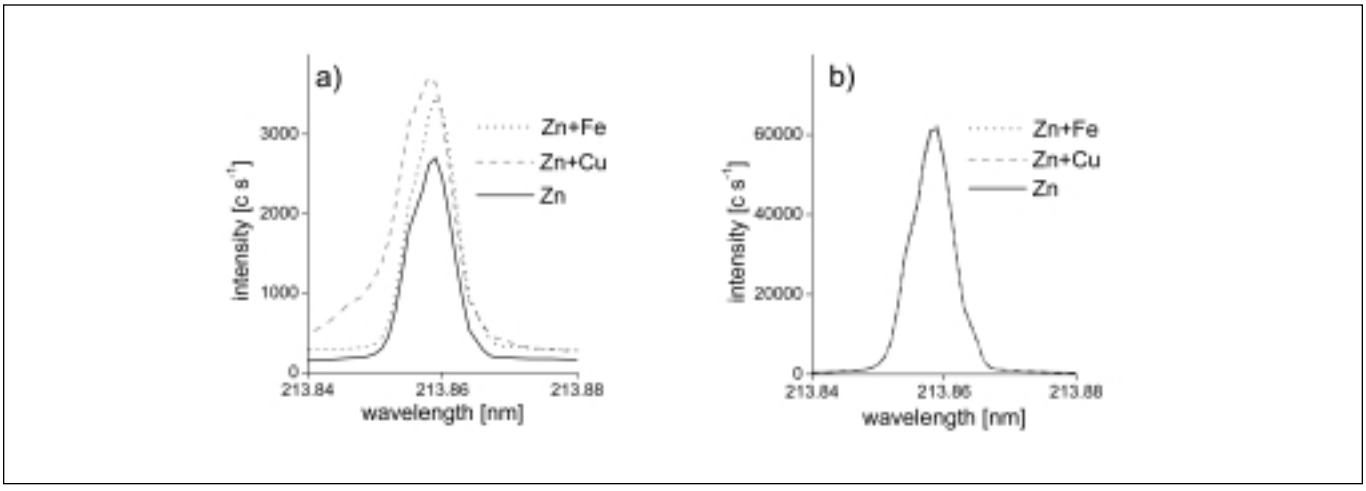


*Fig. 1 (a + b). Zinc signals with and without interference.*
*Figure 1a was measured with an interferent to Zn ratio of 250 (25 mg $L^{-1}$ to 0.1 mg $L^{-1}$).*
*Figure 1b was measured with an interferent to Zn ratio of 1 (2.5 mg $L^{-1}$ to 2.5 mg $L^{-1}$).*

**TABLE VII**
**Limits of Robust Relative Repeatability of Nine Elements Determined From Five Premixes**

| | 1-g Samples (r%) | | | | | 2.5-g Samples (r%) | |
|---|---|---|---|---|---|---|---|
| | A | B | C | D | E | D | E |
| Ca | - | - | 2 | - | - | - | - |
| Cr | - | 4 | - | - | - | - | - |
| Cu | 4 | 3 | 5 | 11 | 37 | 13 | 6 |
| Fe | 4 | 4 | 5 | 9 | 20 | 13 | 23 |
| Mg | - | - | - | - | 17 | - | 9 |
| Mn | 5 | 3 | - | 10 | 39 | 12 | 16 |
| Mo | - | 5 | - | - | - | - | - |
| Se | - | 4 | - | 84 | - | 34 | - |
| Zn | 5 | 4 | 4 | 10 | 37 | 9 | 16 |

**TABLE VIII**
**Limits of Robust (Relative) Intermediate Reproducibility of Nine Elements Determined From Five Premixes**

| | 1-g Samples (iR%) | | | | | 2.5-g Samples (iR%) | |
|---|---|---|---|---|---|---|---|
| | A | B | C | D | E | D | E |
| Ca | - | - | 4 | - | - | - | - |
| Cr | - | 16 | - | - | - | - | - |
| Cu | 5 | 12 | 5 | 13 | 59 | 7 | 44 |
| Fe | 5 | 8 | 9 | 26 | 36 | 14 | 15 |
| Mg | - | - | - | - | 19 | - | 22 |
| Mn | 6 | 8 | - | 14 | 38 | 9 | 31 |
| Mo | - | 8 | - | - | - | - | - |
| Se | - | 5 | - | 73 | - | 33 | - |
| Zn | 3 | 4 | 6 | 12 | 63 | 9 | 48 |

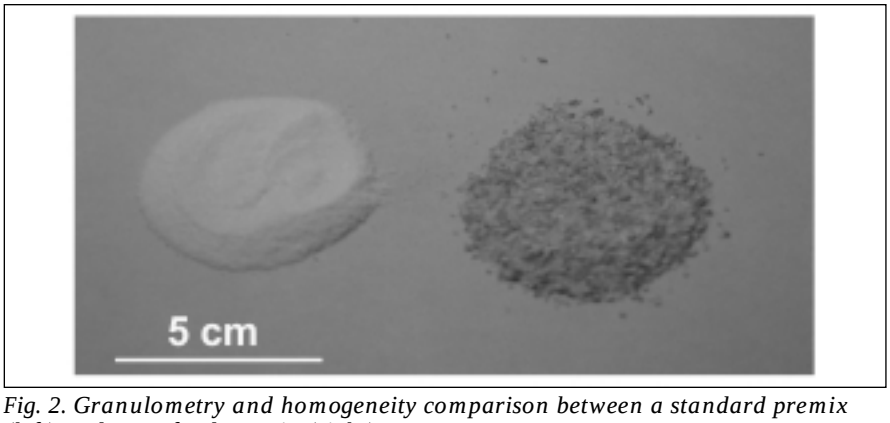


*Fig. 2. Granulometry and homogeneity comparison between a standard premix (left) and a pet food premix (right).*

The reason for the high repeatability limits of premixes D and E is explained by the heterogeneity of the materials as shown in Figure 2 [photograph of a standard premix (left) and a pet food premix (right)]. It is recommended to use 2.5 g as the sample size for pet food premixes. A homogenization of the pet food premixes by grinding a larger sample amount (>50 g) would certainly improve the repeatability of the method for the premix samples. However, two points have to be considered when applying grinding as a sample pretreatment:

(a) The gained homogeneity of the sample does not reflect the actual heterogeneity of the premix (loss of information about the quality of the premix).

(b) Homogeneity is harder to achieve for pet food premixes (based on salts as the major constituent) than for standard premixes (based on maltodextrine).

**Accuracy**

Due to a lack of certified reference materials for premix analysis, accuracy of the method was determined by the following steps:

(a) Comparison between the manufacturer's declared values and measured values, and

(b) By spiking recoveries.

(a) The first approach can only account for a qualitative estimation of accuracy of the method as the manufacturer's declared values are only based on the weights of different salts mixed together during the premix production. Heterogeneity of a production lot cannot be estimated without exactly knowing the production process (quantity and grain size of the different components). The manufacturer's declared and measured concentrations are listed in Table IX. The medians of the measured concentrations were obtained from the

**TABLE IX. Manufacturer's Declared (Given) and Actually Measured Concentrations in Five Premixes**

**(Measured concentrations are medians of 14 replicates. Values in bold letters are measured concentrations of the given concentration range.)**

| | | Concentration (mg kg$^{-1}$) Premix | | | | |
|---|---|---|---|---|---|---|
| | | A | B | C | D | E |
| Ca | Given | - | - | 62500-76400 | - | - |
| | Measured | | | **67650** | | |
| Cr | Given | - | 77-85 | - | - | - |
| | Measured | | **85** | | | |
| Cu | Given | 460-531 | 3600-3960 | 51-59 | 3750-4500 | 433-520 |
| | Measured | **502** | **3790** | **55** | **4208** | **459** |
| Fe | Given | 9420-10870 | 27500-29700 | 510-590 | 25100-30200 | 3460-4100 |
| | Measured | **9980** | **28380** | **488** | **25400** | **3615** |
| Mg | Given | - | - | - | - | 25000-30000 |
| | Measured | | | | | 23900 |
| Mn | Given | 34-38 | 6540-13080 | - | 2050-2500 | 660-790 |
| | Measured | 43 | **7300** | | 3688 | 840 |
| Mo | Given | - | 214-235 | - | - | - |
| | Measured | | 208 | | | |
| Se | Given | - | 69-77 | - | 44-54 | - |
| | Measured | | **71** | | 41 | |
| Zn | Given | 5320-6140 | 32000-35200 | 670-780 | 49600-59500 | 3430-4100 |
| | Measured | **5640** | **32700** | **736** | 47601 | 4140 |

**TABLE X. Recoveries Obtained From Spikes at Two Levels for Premixes A–C and One Level for Premixes D–E**

**(Recoveries of premixes A–C and of D–E were obtained from 1-g and 2.5-g sample sizes, respectively. Values are means of duplicates ± half the range between duplicates.)**

| Premix | A | | B | | C | | D | E |
|---|---|---|---|---|---|---|---|---|
| Spike Conc | 100% | 20% | 100% | 20% | 100% | 20% | 100% | 100% |
| Analyte | Spike Recovery (%) | | | | | | | |
| Ca | - | - | - | - | 95 ± 1 | 90 ± 7 | - | - |
| Cr | - | - | 93 ± 1 | 94 ± 4 | - | - | - | - |
| Cu | 103 ± 3 | 87 ± 1 | 100 ± 2 | 120 ± 25 | 94 ± 1 | 88 ± 6 | 96 ± 2 | 101 ± 1 |
| Fe | 98 ± 1 | 89 ± 4 | 96 ± 1 | 80 ± 5 | 96 ± 1 | 89 ± 7 | 88 ± 6 | 104 ± 1 |
| Mg | - | - | - | - | - | - | - | 91 ± 8 |
| Mn | 93 ± 2 | 81 ± 5 | 97 ± 1 | 113 ± 16 | - | - | 92 ± 4 | 96 ± 2 |
| Mo | - | - | 99 ± 1 | 103 ± 3 | - | - | - | - |
| Se | - | - | 100 ± 2 | 100 ± 2 | - | - | 124 ± 12 | - |
| Zn | 104 ± 2 | 93 ± 6 | 95 ± 1 | 81 ± 2 | 97 ± 1 | 84 ± 8 | 94 ± 2 | 102 ± 1 |

repeatability determinations (1-g sample size and 14 replicates for each premix). Sixteen of the 25 measured concentrations (values given in bold letters in Table IX) were in the range of the manufacturer's declaration. The values found out of the declaration range were not very far from the supplier's indications, which could be due to an inaccurate sampling procedure.

(b) Premixes A–E were spiked before digestion by pipetting 1 mL and 2.5 mL of concentrated solutions into premixes A–C (1-g samples) and D–E (2.5-g samples), respectively (see Table X). By applying small volumes of concentrated solutions, it was intended to minimize the dilution of the added acid and its digestion efficiency. Two levels of spiking were applied, corresponding to 20 and 100% of the concentrations of the analyte obtained from the sample. Only elements with claimed concentrations were taken into account.

For most of the 100% level spikes, the recoveries for the five investigated premixes (except Se in D) were satisfactory. For the 20% spikes (premixes A–C), the recoveries ranged from 80–120%, which can be considered satisfactory because good recoveries in low spikes are more difficult to achieve. For premixes D–E, the results for the 20% spikes were disregarded because of the heterogeneity of the original sample. Considering a variability of ±10% for the original sample concentration and the same variability for the spiked sample, the worst case recoveries could be estimated to be between 0 and 200%.

## CONCLUSION

A method for the simultaneous determination of Ca, Cr, Cu, Fe, Mg, Mn, Mo, Se, and Zn in mineral premixes using ICP-OES has been evaluated. These food additives feature high concentration ranges of minerals and therefore demand high sensitivity and low interference for low mineral content and high dilution factors for high mineral content in premixes. The linearity of calibration was good for eight elements, only Mg had a quadratic calibration curve. Commonly used wavelengths for the nine elements were checked for interferences, but none was found significant when measured under premix conditions. The limits of quantitation were below the concentrations expected for the studied premixes, except for Se, where the lowest required concentration was half the limit of quantitation.

Robust relative repeatability and robust intermediate reproducibility were < 5% and < 16%, respectively, for three premixes out of five. Two other premixes were found to be very heterogeneous and were analyzed using a larger sample size. The robust relative repeatability for these samples was between 6% and 34% and the intermediate reproducibility was between 7% and 48% depending on the elements and their concentration levels. The recoveries of spikes on the homogeneous samples were relatively good when high spikes were applied (93–104%); the recoveries on the heterogeneous samples were poor (88–124%). The developed multielement method performed well on homogeneous premixes; however, high repeatability data are to be expected from pet food premixes due to their high heterogeneity.

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## REFERENCES

1. L.A. Mejia, Food and Nutrition Bulletin 19, 278 (1995).
2. C. Reilly, Trends in Food Science and Technology 9, 114 (1998).
3. J.S. Kosse, A.C. Yeung, A.I. Gil, and D.D. Miller, Food Chemistry 75, 371 (2001).
4. C.P. Hanna, G.R. Carnrick, S.A. MacIntosh, L.C. Guyette, and D.E. Bergemann, At. Spectrosc. 16, 82 (1995).
5. L. Valiente, M. Piccinna, E.R. Ale, A. Grillo, and P. Smichowski, At. Spectrosc. 23, 129 (2002).
6. C.P. Bosnak and K.W. Barnes, At. Spectrosc. 19(2), 40 (1998).
7. International Organization for Standardization, International Standard ISO 6869:1 (2000).
8. L. Jorhem, Journal of AOAC International 83, 1204 (2000).
9. L. Jorhem and J. Engman, Journal of AOAC International 83, 1189 (2000).
10. N.J. Miller-Ihli, J. Agric. Food Chem. 44, 2675 (1996).
11. P.D. Krampitz and K.W. Barnes, At. Spectrosc. 19, 43 (1998).
12. K. Mitko and M. Bebek, At. Spectrosc. 20, 217 (1999).
13. S.P. Dolan and S.G. Capar, Journal of Food Composition and Analysis 15, 593 (2002).
14. A. Moeller, R.F. Ambrose, and S.S. Que Hee, Food Additives and Contaminants 18, 19 (2001).
15. D-H. Sun, J.K. Waters, and T.P. Mawhinney, Journal of AOAC International 83, 1218 (2000).
16. L. Perring and M. Basic-Dvorzak, Anal. and Bioanal. Chem. 374, 235 (2002).