

Supplementary Information

Voltammetric Electronic Tongue for Discrimination of Milk Adulterated with Urea, Formaldehyde, and Melamine *Chemosensors* 2014, 2, 251-266

**L ígia Bueno [†], William R. de Araujo [†], Maiara O. Salles [†], Marcos Y. Kussuda
and Thiago R. L. C. Paix ão ***

Instituto de Química, Universidade de São Paulo, São Paulo 05508-000, SP, Brazil;
E-Mails: ligia-b@usp.br (L.B.); william_reis_araujo@hotmail.com (W.R.A.);
maiara_os@yahoo.com.br (M.O.S.); marcosy@iq.usp.br (M.Y.K.)

[†] These authors contributed equally to this work.

* Author to whom correspondence should be addressed; E-Mail: trlcp@iq.usp.br;
Tel.: +55-11-3091-9150.

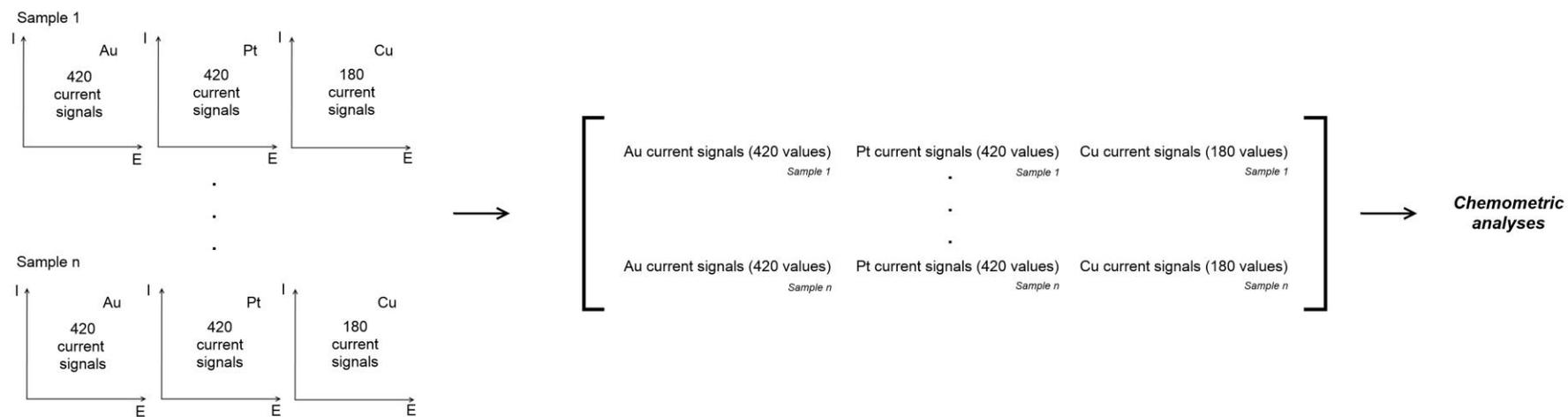
Scheme S1. Schematic illustration of how the current signals were organized in order to use the Statistica software.

Figure S1. Cyclic voltammograms recorded directly in skimmed milk using platinum (A), gold (B), and copper (C) working electrodes, in the absence (dashed lines) and presence (full lines) of 10.0 mmol L⁻¹ of formaldehyde (1), 0.95 mmol L⁻¹ of melamine (2), and 4.16 mmol L⁻¹ of urea (3). Scan rate = 100 mV s⁻¹.

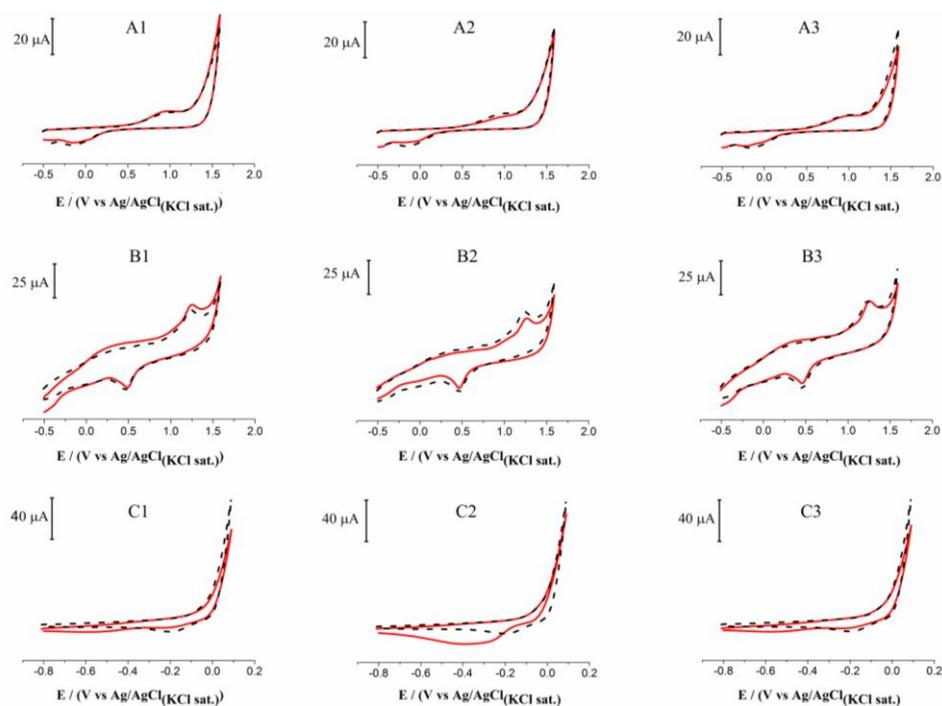


Figure S2. Cyclic voltammograms recorded directly in semi-skimmed milk using platinum (A), gold (B), and copper (C) working electrodes, in the absence (dashed lines) and presence (full lines) of 10.0 mmol L⁻¹ of formaldehyde (1), 0.95 mmol L⁻¹ of melamine (2), and 4.16 mmol L⁻¹ of urea (3). Scan rate = 100 mV s⁻¹.

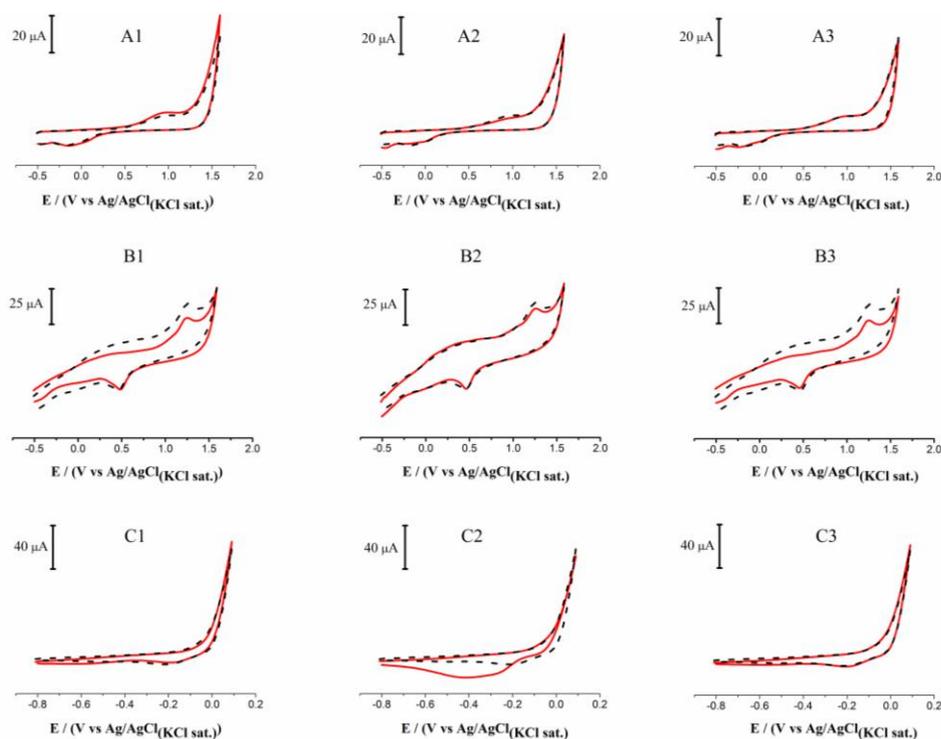


Figure S3. (A) Voltammogram recorded in 0.10 mol L⁻¹ acetate buffer (pH 4.5) (full line) after the addition of formaldehyde (dashed line), melamine (dotted line), and urea (dashed dotted line) at a final concentration of 9.5 mmol L⁻¹. Scan rate = 50 mV s⁻¹. (B) Frequency shifts versus potential plots for gold quartz crystal registered at the same time as the voltammograms.

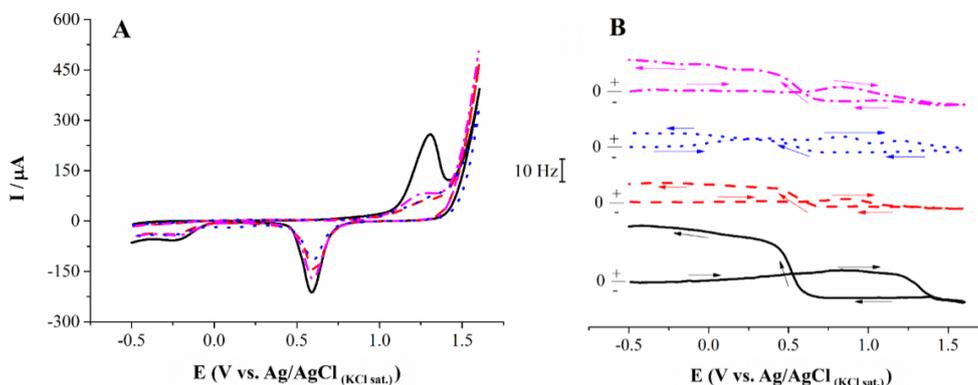


Figure S4. The HCA algorithm and Euclidian distances using whole milk data extracted using gold (A), copper (B), and platinum (C) electrodes with a Ag/AgCl(KCl sat) reference electrode. Samples were analysed in triplicate.

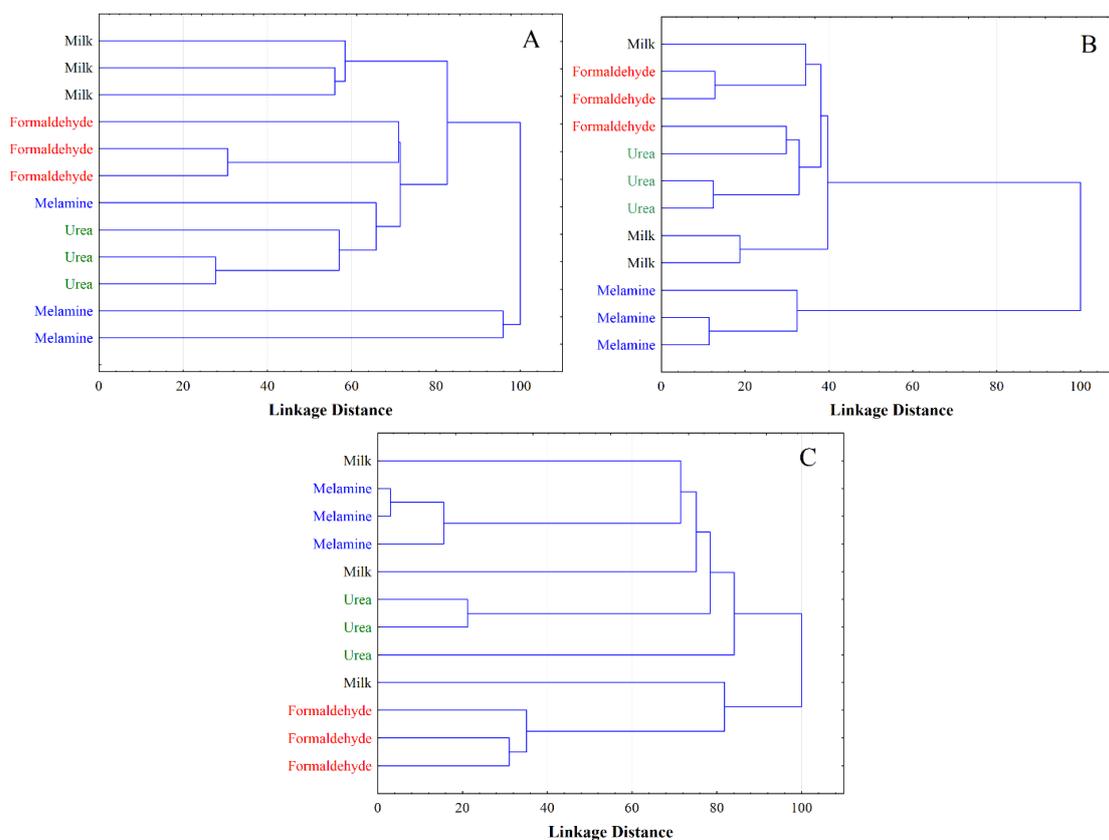


Figure S5. The HCA algorithm and Euclidian distances using data extracted from commercial (Elegê®) (RF S.A.–Entrepósito Usina, Teutônia, RS, Brazil) whole milk samples using three working electrodes (gold, platinum, and copper) with a Ag/AgCl_(KCl sat) reference electrode. Samples were analysed in triplicate.

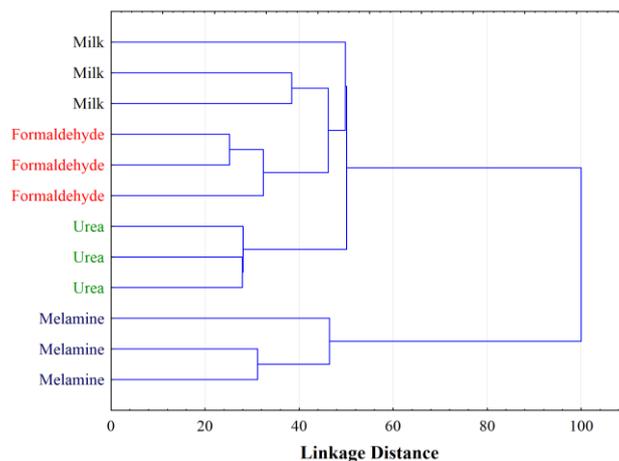


Figure S6. The PCA loading plot [tri-dimensional view (A) and top view (B)] using the input data reported in Figure. 4. Symbols: Au (red circles), Pt (green circles) and Cu (blue circles).

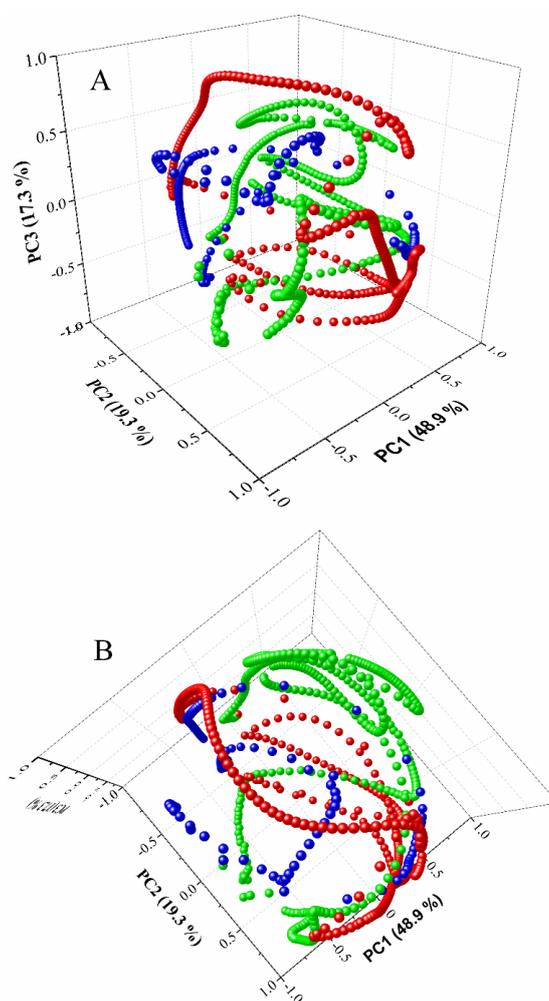


Figure S7. PCA 3D plots of commercial (Parmalat®) (Laticínios Bom Gosto S.A. Usina de Beneficiamento, Tapejara, RS, Brazil) milk samples: unadulterated samples (black squares), and samples adulterated with 10.0 mmol L⁻¹ of formaldehyde (red circles), 0.95 mmol L⁻¹ of melamine (blue up triangles), and 4.16 mmol L⁻¹ of urea (green down triangles). Whole milk (A), skimmed milk (B), and semi-skimmed milk (C). Samples were analysed in triplicate.

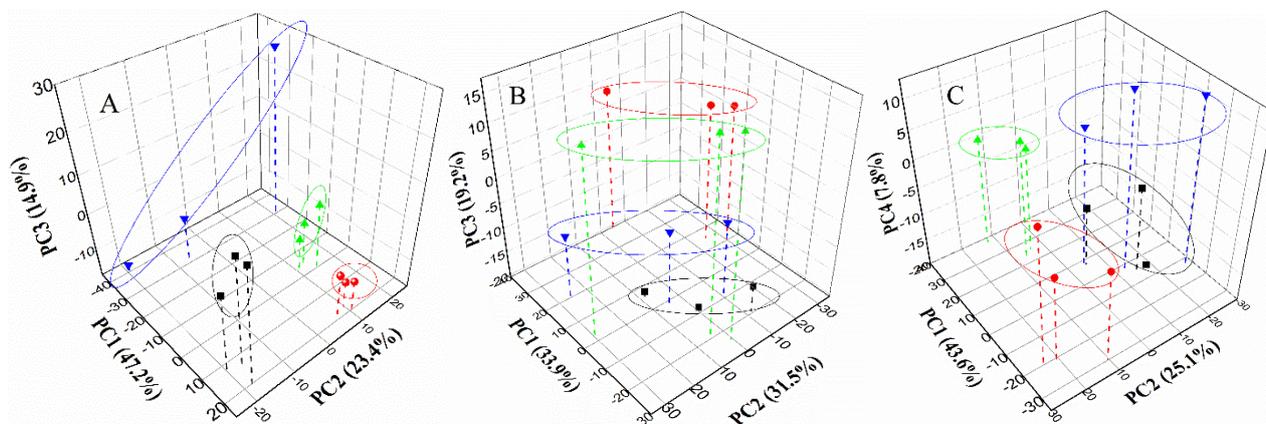


Figure S8. PCA 3D plots of commercial (Qualitã®) (Usina de Beneficiamento de Leite da Cooperativa dos Suinocultores de Encantado Ltda, Arroio do Meio, RS, Brazil) milk samples: unadulterated samples (black squares), and samples adulterated with 10.0 mmol L⁻¹ of formaldehyde (red circles), 0.95 mmol L⁻¹ of melamine (blue up triangles), and 4.16 mmol L⁻¹ of urea (green down triangles). Whole milk (A), skimmed milk (B), and semi-skimmed milk (C). Samples were analysed in triplicate.

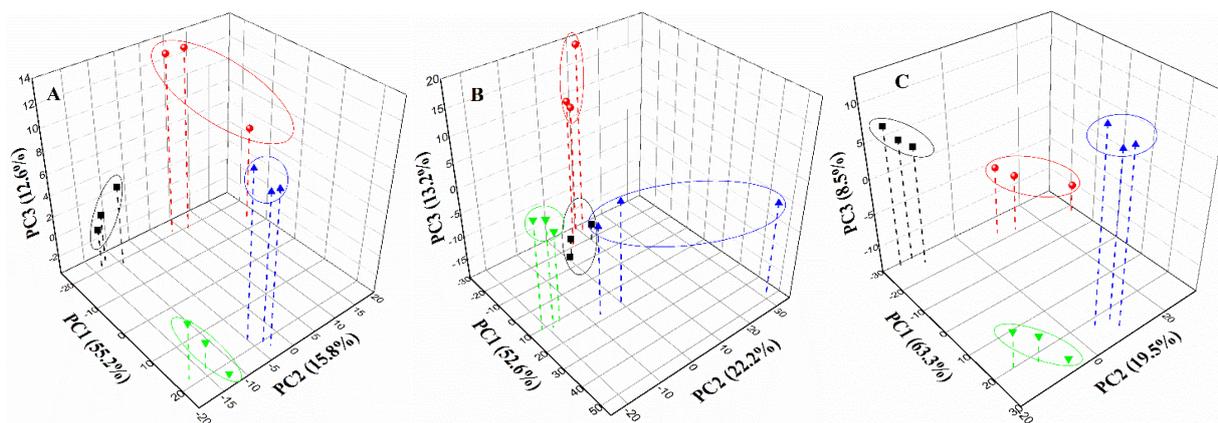


Figure S9. PCA 3D plots of commercial (Elegê®) (BRF S.A. – Entrepósito Usina, Teutônia, RS, Brazil) whole milk samples: unadulterated samples (black squares), and samples adulterated with 100 mmol L^{-1} of formaldehyde (red circles), 9.5 mmol L^{-1} of melamine (blue up triangles), and 41.6 mmol L^{-1} of urea (green down triangles). Samples were analysed in triplicate.

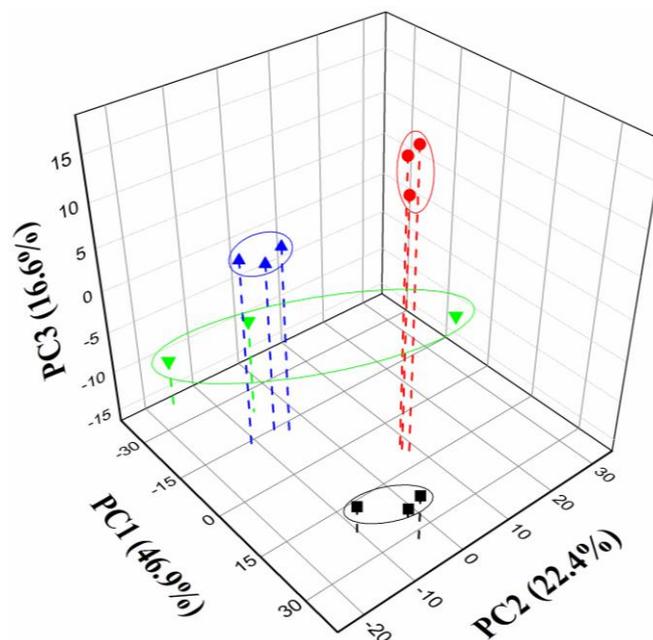


Figure S10. PCA 3D plots of commercial (Elegê®) (BRF S.A.–Entrepósito Usina, Teutônia, RS, Brazil) whole milk sample: unadulterated samples (black squares), and samples adulterated with 10 mmol L^{-1} (red circles) and 100 mmol L^{-1} (open red circles) of formaldehyde; 0.95 mmol L^{-1} (blue up triangles) and 9.5 mmol L^{-1} (open blue up triangles) of melamine; and 4.16 mmol L^{-1} (green down triangles) and 41.6 mmol L^{-1} (open green down triangles) of urea. Samples were analysed in triplicate.

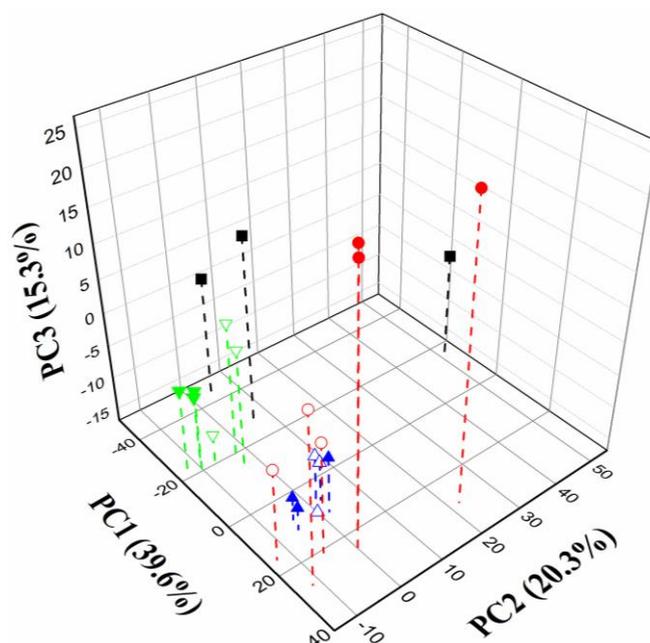
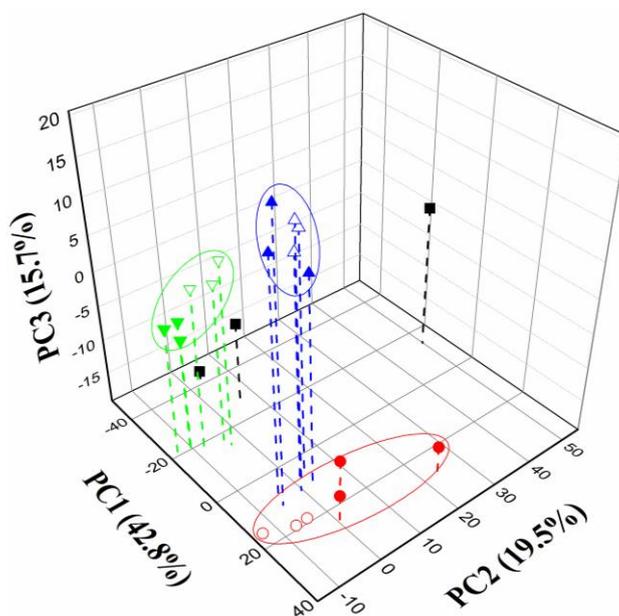


Figure S11. PCA 3D plots of commercial (Eleg[®]) (BRF S.A.–Entrepoto Usina, Teutônia, RS, Brazil) whole milk samples: unadulterated samples (black squares), and samples adulterated with 10 mmol L⁻¹ (red circles) and 100 mmol L⁻¹ (open red circles) of formaldehyde; 0.95 mmol L⁻¹ (blue up triangles) and 9.5 mmol L⁻¹ (open blue up triangles) of melamine; and 4.16 mmol L⁻¹ (green down triangles) and 41.6 mmol L⁻¹ (open green down triangles) of urea. Samples were analysed in triplicate. All the input data was pre-treated using a local technique transformation reported in the manuscript.



© 2014 by the authors; licensee Molecular Diversity Preservation International, Basel, Switzerland. This article is an open-access article distributed under the terms and conditions of the Creative Commons Attribution license (<http://creativecommons.org/licenses/by/4.0/>).