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ARTICLE

Three-Year Comparison of the Polyphenol Contents and Antioxidant Capacities in Organically and Conventionally Produced Apples (*Malus domestica* Bork. Cultivar 'Golden Delicious')

Berenike A. Stracke,[†] Corinna E. Rüfer,[†] Franco P. Weibel,[§] Achim Bub,[†] and Bernhard Watzl*,[†]

[†]Department of Physiology and Biochemistry of Nutrition, Max Rubner-Institute, Federal Research Institute of Nutrition and Food, Karlsruhe, Germany, and [§]Research Institute of Organic Agriculture, Frick, Switzerland

The present study was performed to evaluate the polyphenol content and antioxidant capacity of apples (cv. 'Golden Delicious') grown under defined organic and conventional conditions. Apples were harvested at five comparable commercial farms over the course of three years (2004−2006). In 2005 and 2006 the antioxidant capacity was 15% higher ($p < 0.05$) in organically produced apples than in conventionally produced fruits. In 2005 significantly higher polyphenol concentrations were found in the organically grown apples. In 2004 and 2006 no significant differences were observed (2004, 304 ± 68 μg/g organic vs 284 ± 69 μg/g conventional, $p = 0.18$; 2005, 302 ± 58 μg/g organic vs 253 ± 41 μg/g conventional, $p = 0.002$; 2006, 402 ± 100 μg/g organic vs 365 ± 58 μg/g conventional, $p = 0.17$). Year-to-year variations in the antioxidant capacity and the polyphenol content of up to 20% were more significant than the production method found within one year. Finally, flavanols and flavonols were major determinants of the antioxidant capacities in these apples. Overall, the production method had a smaller impact on the variation in the polyphenol content and antioxidant capacity of apples than the yearly climate.

KEYWORDS: Organic; apples; phytochemicals; antioxidant capacity

INTRODUCTION

To date approximately 8% of the agricultural area in European countries is managed organically (*1*). In Germany, organic agriculture increased by about 30% over the past 10 years (*1*). This development is in line with a change in consumer behavior, on the discussion about safer and healthier food, for example, which contains less pesticide residues. More consumers purchase organic products at the supermarket or specialty stores and expect healthier foods (*2*). Therefore, two questions arise: Which compounds in foods contribute to their health value? Are there significant differences in the content of these compounds between organically and conventionally produced foods?

Epidemiological studies have linked apple consumption to a reduced risk of lung cancer and asthma (*3*, *4*). Furthermore, a human intervention study showed that apple consumption decreased levels of endonuclease III sensitive sides and increased protection against DNA damage induced by iron chloride (*5*). In animal studies cloudy apple juice inhibited DNA damage and colon carcinogenesis induced by 1,2-dimethylhydrazine (*6*, *7*).

*Address correspondence to this author at the Department of Physiology and Biochemistry of Nutrition, Max Rubner-Institute, Federal Research Institute of Nutrition and Food, Haid-and-Neu-Str. 9, 76131 Karlsruhe, Germany (telephone: +49 721 6625 410; fax: +49 721 6625 404; e-mail: bernhard.watzl@mri.bund.de).

These associations are partly linked to the high content of phytochemicals (e.g., flavonoids) in apples (*8*). Apples contain about 2 g of polyphenols/kg of fresh weight, depending on the cultivar. They are rich in flavonoids (e.g., flavanols, flavonols, dihydrochalcones) and contain high amounts of hydroxycinnamic acid derivates, mainly chlorogenic acid (*9*). Some phytochemicals (e.g., flavonoids) exhibit stronger in vitro antioxidant capacities than classic antioxidant vitamins such as vitamins C and E (*10*).

The content and profile of phenolic compounds in plants are influenced by several factors, for instance, the exposure to pathogens when pesticide use is avoided (*11*, *12*), climate, crop variety, and degree of ripeness (*2*, *13*). Studies have shown that some herbicides reduce the carbon fixation of plants, thus decreasing the proportion of carbon available for the synthesis of secondary metabolites (*14*, *15*). However, data on the influence of the production method on the content of phenolic compounds and the antioxidant capacity in apples are inconsistent. In 2000 and 2004 Weibel et al. reported an 18−23% higher content of phenolic compounds in organically produced apples (cv. 'Golden Delicious') (*16*, *17*). In contrast, two other studies reported no differences in the phenolic contents between organically and conventionally produced apples (*18*, *19*). Chinnici et al. and Tarozzi et al. were able to show that integrated apples have a higher antioxidant capacity than organically produced

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apples (*18*, *20*). These data indicate that the biosynthesis of phenolic compounds depends not only on the agricultural systems but also on other less characterized and controlled factors such as climate. Therefore, to compare different agricultural systems it is important that the respective fruits and vegetables are grown, harvested, and stored under comparable and well-defined conditions (e.g., soil conditions, climate, cultivar, stage of ripeness). Unfortunately, this information is not always stated in the cited studies and may explain the various results.

The aim of the present study was to determine the antioxidant capacity and the polyphenol content (hydroxycinnamic acids, dihydrochalcones, flavanols, and flavonols) of apples (cv. 'Golden Delicious'). They were grown under well-defined organic and conventional conditions for a period of 3 years and represent a consumer-relevant range of production regions and sites.

MATERIALS AND METHODS

Chemicals. Chlorogenic acid, phloretin 2′-xyloglucoside, phloretin 2′-glucoside (phloridzin), catechin, and epicatechin were purchased from Fluka (Taufkirchen, Germany), and procyanidin B1 and B2 were from LGC Promochem GmbH (Wesel, Germany). Quercetin and quercetin 3-rhamnoside were obtained from Roth (Karlsruhe, Germany). 2,2′-Azinobis(3-ethylbenzothiazoline-6-sulfonic acid) diammonium salt (ABTS) was purchased from Sigma (Munich, Germany), α,α′-azodiisobutyramidine dihydrochloride (ABAP) and 2,4,6-tri-2-pyridyl-1,3,5-triazine (TPTZ) as well as fluorescein were also obtained from Fluka. Unless otherwise stated, all other chemicals were purchased from Merck (Darmstadt, Germany).

Apple Production. From an existing network for system comparison studies in Switzerland, which includes 10 commercial farms (5 comparison pairs), organic and conventional (integrated) apples of the cv. 'Golden Delicious" were originated (*16*). At all five sites there are neighboring commercial orchards with certified organic and conventional productions, respectively. All organic orchards were under certified organic management for more than 4 years. Organic matter content of the 0−25 cm layer per orchard was assessed (titration method, data not shown). The proximity between the organic and conventional fields varied from 500 m to 2 km. Crop year differences were analyzed by three of five neighboring commercial farms pairs for three years (2004, 2005, and 2006). The organically and conventionally grown apples were cultivated and certified according to the regulations of "Bio Suisse" (organic) and "Suisse Garantie" (certified integrated production, termed "conventional" in the following). Orchard, soil, and microclimate conditions per farm pair were comparable during the three years. Plant protection, fertilization, and crop load regulation were recorded in detail. Each year the same products and concepts were used. Soil analyses were carried out for 2004 (soil characteristics are reported in **Table 1**). The analyzed apples were sampled by scientists of the Swiss Research Institute of Organic Agriculture (FiBL) according to a detailed protocol: 50 kg or 350 fruits of each orchard were harvested at the first picking passage from trees with a representative crop load and at optimal maturity (determinated by the Streif index). Two representatively developed fruits (fulfilling table fruit quality standards) were sampled per tree from the middle canopy zone. The apples were immediately transported to the same cold-storage room at FiBL (2 °C and 93% relative humidity). The apples were harvested in 2004, 2005, and 2006.

Standard Quality of Apples. Fruit weight, fruit flesh firmness, ripening index, mineral element, and sugar content (°Brix) were measured to quantify the standard quality of apples (*16*).

Analysis of Dry Matter in Apples. Approximately 5 g of the homogenates was dried for 5 h at 110 °C. The analysis was repeated five times.

Extraction of Polyphenols. Six apples were randomly chosen from the organic and conventional samples. Apples were sliced into fourths, seeds and core were removed, and one-fourth of each apple was homogenized using a laboratory blender. Approximately 3 g of the homogenate was extracted with 10 mL of methanol in an ultrasonic bath for 60 min at 50 °C. The samples were centrifuged for 10 min at 3000*g*. The supernatant was filtered through a filter paper (no. 595, Schleicher & Schell, Dassel, Germany) using a funnel. After three washings of the pellet with 5 mL of methanol, the residue was filtered again. The combined organic phases were evaporated using a rotary evaporator Laborota 4003-digital (Heidolph, Schwabach, Germany). The remaining aqueous residue was filled to 5 mL with distilled water for HPLC analysis and antioxidant tests. The extraction was repeated five times.

Table 1. Soil Characteristics for Organically and Conventionally Produced Apples in Five Comparable Farm Pairs (Year 2004)[a]

	organic	conventional
organic matter (%)	2.5 ± 1.2 (1.3−4.5)	3.1 ± 1.6 (1.4−5.9)
phosphorus (mg/kg)	185.6 ± 74.5 (98.1−334.7)	144.1 ± 41.2 (88.8−225.5)
potassium (mg/kg)	266.8 ± 211.0 (43.9−612.0)	210.9 ± 83.6 (131.1−365.2)

[a] Values are means ± SD. The minimum and maximum values are shown in parentheses. No significant differences were observed between the organic and conventional soil characteristics (extraction methods, P and K, ammonium-EDTA; organic matter, titration method).

HPLC Analysis of Phenolic Compounds. HPLC analysis was performed with a high-pressure gradient system from Shimadzu (Duisburg, Germany) equipped with an autoinjector, a photodiode array detector, and a fluorescence detector. Separation was carried out with a Prontosil (250 mm × 4.6 mm i.d., particle size = 3 μm) reversed-phase column (Bischoff, Leonberg, Germany). Solvent A consisted of 0.1% formic acid in water (pH 3) and solvent B of acetonitrile. A linear gradient was used: from 15 to 30% B in 50 min, from 30 to 50% B in 10 min, and from 50 to 56.5% B in 20 min. For analysis of the flavanols catechin and epicatechin elution was effected using the following linear gradient: 15% B for 25 min, from 15 to 30% B in 25 min, and from 30 to 50% B in 10 min. The flow rate was set to 0.8 mL/min, and the injection volume was 50 μL. The eluent was recorded with diode array detection at 280 nm for quantification of the flavanols procyanidin B1 and B2, at 290 nm for the dihydrochalcones, at 320 nm for the hydroxycinnamic acids, and at 350 nm for the flavonols. Peaks were scanned between 190 and 500 nm. For quantification of catechin and epicatechin the fluorescence detection settings were an excitation wavelength of 280 nm and an emission wavelength of 320 nm.

Quantification was performed by external calibration using commercially available reference compounds. Calibration curves for the different polyphenols were in the range of 0.1−100 μM in which the linearity of the response was given. Quantification of those polyphenols that were not commercially available was based on a representative standard of the same polyphenol class. The recovery for all polyphenols was >95% obtained after spiking apple samples with polyphenols and subtracting the basal values from the blank samples. The coefficient of variation of the method was below 10% (intra-assay). The limits of detection ranged from 200 fmol to 5 pmol.

For identification of the polyphenols without commercially available reference compounds, a HPLC-MS analysis was performed with a HP 1100 series HPLC (Agilent Technologies, Waldbronn, Germany) equipped with an autoinjector, a binary HPLC pump, a column heater, a UV detector, and a HP Chem Station for data collection and handling. The HPLC was interfaced to a HP series 1100 mass selective detector equipped with an atmospheric pressure ionization−electrospray (API-ES) chamber. The apple polyphenols were analyzed under the following conditions: capillary voltage, 3.5 kV; fragmentor voltage, 150 V; nebulizing pressure, 50 psi; drying gas temperature, 350 °C; and drying gas flow, 12.5 L/min. The scan mode was used for data collection. Spectra were scanned over a mass range of m/z 100−600 at 0.98 s per cycle. For HPLC analysis the same conditions as described above were used.

Antioxidant Capacity. The antioxidant capacity was determined using three different antioxidant assays. Procedures followed were similar to those described by Benzie and Strain (*21*) for the ferric reducing antioxidant power (FRAP) assay, Cao and Prior (*22*) for the oxygen radical antioxidant capacity (ORAC) assay, and Re (*23*) for the Trolox equivalent antioxidant capacity (TEAC) assay.

FRAP Assay. The reaction was performed in 300 mM acetate buffer (pH 3.6), and the final reaction mixture was 340 μL. Trolox (50, 100, 150, 250, or 300 μM; used as standard), apple samples, or buffer alone (blank) (10 μL each) was mixed with 30 μL of distilled water in 96-well plates

Table 2. Antioxidant Capacity in Apples (Cv. 'Golden Delicious') from Organic (org) and Conventional (conv) Production Methods in 2004 (**A**), 2005 (**B**), and 2006 (**C**)[a]

		FRAP (μmol TE/g FW)	ORAC (μmol TE/g FW)	TEAC (μmol TE/g FW)
		(**A**) 2004		
FP1	org	4.1 ± 1.1	9.0 ± 1.3	7.9 ± 2.2
	conv	4.5 ± 1.1	7.9 ± 1.5	8.8 ± 2.3
FP2	org	4.5 ± 1.2 a	8.8 ± 1.6	8.6 ± 2.5 a
	conv	4.2 ± 0.8	8.0 ± 1.0	8.0 ± 1.5
FP3	org	5.1 ± 0.9 a	9.2 ± 1.3	9.6 ± 1.5 a
	conv	3.7 ± 0.9	8.3 ± 0.9	7.0 ± 1.8
FP4	org	3.8 ± 0.6 a	8.6 ± 1.2	7.6 ± 1.1 a
	conv	3.2 ± 0.6	8.2 ± 1.0	6.3 ± 1.0
FP5	org	2.9 ± 0.6	7.2 ± 1.2	6.0 ± 1.2
	conv	3.7 ± 0.7	7.4 ± 0.7	7.4 ± 1.4
mean	org	4.1 ± 1.1	8.6 ± 1.4	7.9 ± 2.1
	conv	3.9 ± 0.9	7.9 ± 1.0	7.6 ± 1.8
		(**B**) 2005		
FP1	org	3.6 ± 0.5	5.9 ± 0.4	6.5 ± 0.7
	conv	4.1 ± 0.1	6.0 ± 0.3	7.3 ± 0.2 b
FP2	org	3.9 ± 0.7 a	7.2 ± 1.2 a	7.4 ± 1.3 a
	conv	3.4 ± 0.4	6.1 ± 0.7	6.2 ± 0.9
FP3	org	4.3 ± 0.6 a	8.8 ± 1.2 a	8.1 ± 1.1 a
	conv	3.5 ± 0.5	7.7 ± 0.7	6.6 ± 1.1
FP4	org	3.8 ± 0.4 a	6.5 ± 0.8 a	7.3 ± 0.8 a
	conv	3.4 ± 0.5	5.0 ± 0.9	6.5 ± 0.9
FP5	org	4.6 ± 0.6 a	9.4 ± 1.5 a	8.8 ± 0.9 a
	conv	3.3 ± 0.5	5.3 ± 0.3	6.1 ± 0.7
mean	org	4.1 ± 0.6 a	7.6 ± 1.7 a	7.6 ± 1.2 a
	conv	3.5 ± 0.5	6.0 ± 1.1	6.5 ± 0.8
		(**C**) 2006		
FP1	org	4.9 ± 0.7 a	8.1 ± 1.6	9.6 ± 1.5 a
	conv	4.1 ± 0.2	8.3 ± 0.5	7.9 ± 0.3
FP2	org	4.8 ± 0.2 a	11.4 ± 1.4 a	9.3 ± 1.4 a
	conv	3.1 ± 0.5	5.6 ± 1.2	6.3 ± 0.8
FP3	org	3.8 ± 0.2 a	8.1 ± 1.4	6.9 ± 0.5 a
	conv	2.9 ± 0.4	7.2 ± 2.1	5.7 ± 0.8
FP6	org	4.3 ± 0.6 a	8.9 ± 0.8	8.5 ± 1.1 a
	conv	3.9 ± 0.4	9.3 ± 1.9	6.9 ± 2.2
FP7	org	3.4 ± 0.3 a	7.6 ± 1.4	6.0 ± 1.1 a
	conv	3.3 ± 0.3	8.7 ± 1.0	6.9 ± 0.7
mean	org	4.3 ± 0.7 a	8.8 ± 1.8	8.1 ± 1.8 a
	conv	3.5 ± 0.6	7.8 ± 1.9	6.7 ± 1.3

[a] Apples were harvested from five comparable farms pairs (FP) in Switzerland. Values are means ± SD; $n = 5$ per group (a = organic significantly higher than conventional ($p < 0.05$); b = conventional significantly higher than organic ($p < 0.05$); three-way ANOVA; Tukey−Kramer post hoc test); TE, Trolox equivalent; FW, fresh weight.

(Greiner, Frickenhausen, Germany). The reaction was started by adding 300 μL of prewarmed (37 °C) FRAP reagent (mixture of 10 mM TPTZ, 300 mM acetate buffer, 20 mM $FeCl_3$) to each well. The absorbance was determined at $\lambda = 585$ nm (microplate reader; Molecular Devices, Workinham, U.K.) after incubation for 30 min at 37 °C. The microplate was shaken prior to each reading. Extracted apple samples were diluted in distilled water to fit into the concentration range of the Trolox calibration curve. All analyses were performed in triplicate. The Trolox equivalent (TE) concentrations of the apples were calculated by subtracting the blank from samples and standards using a linear calibration curve constructed for each assay. One TE equals the net protection produced by 1 mM Trolox.

ORAC Assay. The test system was conducted in 75 mM phosphate buffer (pH 7.4), and the final reaction mixture was 300 μL. Trolox (20, 70, 100, 150, 200, or 250 μM) in 15 μL of buffer (used as standard), 15 μL of apple samples (in buffer) (see below), or 15 μL of buffer alone (blank) as well as 250 μL of a 100 nM fluorescein solution was mixed in 96-well plates (Greiner) and maintained at 37 °C for 5 min. The oxidation reaction was started by adding 35 μL of 275 mM ABAP to each well. The fluorescence of fluorescein was excited at $\lambda = 495$ nm, and the fluorescence emission was detected at $\lambda = 575$ nm. Using a microplate reader (TECAN Spectra Fluor Plus, Crailsheim, Germany), the decay of the fluorescein fluorescence was monitored every 1 min at 37 °C until the fluorescence of the last reading had declined to < 5% of the first reading. The microplate was shaken prior to each reading. Extracted apple samples were diluted in phosphate buffer in the range of the Trolox calibration curve. All reaction mixtures were prepared in duplicate. For quantification the raw data were exported to an Excel (Microsoft) sheet. After normalization of the antioxidant curves (fluorescence versus time) to the curve of the blank, the area under the fluorescence decay curves (AUC) was calculated. The net AUC corresponding to a sample was calculated by subtracting the AUC of the blank. ORAC values for the apple samples were expressed as TE by using the calibration curve calculated for each assay. A quadratic correlation between the net AUC and concentration was obtained for all calibration curves. One TE equals the net protection produced by 1 mM Trolox.

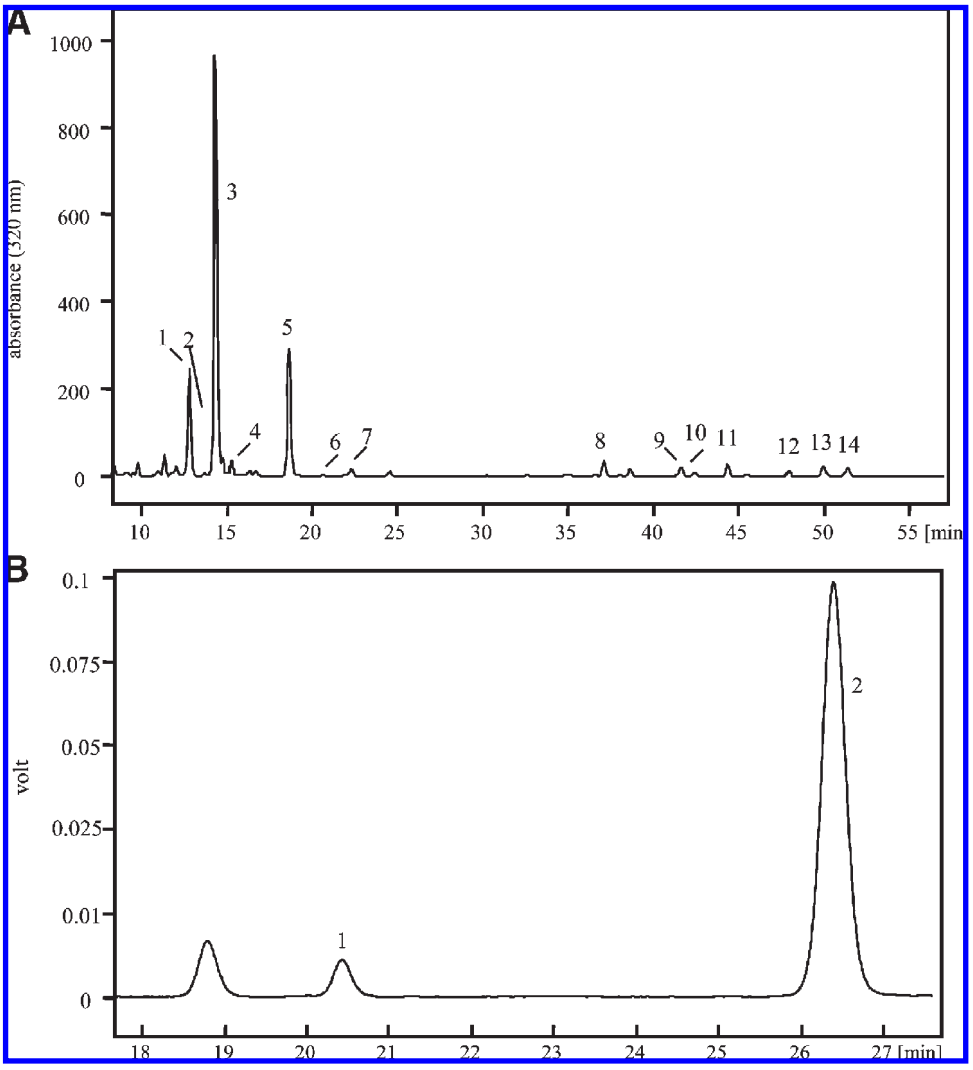

Figure 1. (**A**) Representative HPLC chromatogram of apples (cv. Golden Delicious) at λ = 320 nm: peak 1, 4-caffeoylquinic acid; peak 2, procyanidin B1; peak 3, chlorogenic acid; peak 4, 3-coumaroylquinic acid; peak 5, 4-coumaroylquinic acid; peak 6, procyanidin B2; peak 7, 5-coumaroylquinic acid; peak 8, phloretin 2′-xyloglucoside; peak 9, quercetin 3-galactoside; peak 10, quercetin 3-glucoside; peak 11, phloretin 2′-glucoside; peak 12, quercetin 3-xyloside; peak 13, quercetin 3-arabinoside; peak 14, quercetin 3-rhamnoside. Quantification was performed at different wavelengths (280, 290, 320, 350 nm) as described under Materials and Methods. (**B**) Representative HPLC chromatogram of apples (cv. Golden Delicious) monitored with fluorescence detection at wavelengths 280 nm (excitation) and 320 nm (emission): peak 1, catechin; peak 2, epicatechin

TEAC Assay. The reaction was carried out in 10 mM phosphate buffer (pH 7.4), and the final reaction mixture was 330 μL. Trolox, (50, 100, 150, 200, or 250 μM; used as standard), apple samples (in buffer) (see below), or buffer alone (blank) (30 μL each) was added to 96-well plates (Greiner). The ABTS radical cation ($ABTS^{\bullet+}$) was formed by the reaction of the ABTS stock solution with 2.45 mM potassium persulfate (final concentration) and allowing the mixture to stand in the dark at room temperature for 12−16 h before use. For the assay the $ABTS^{\bullet+}$ solution was diluted with PBS buffer (pH 7.4, 10 mM) to an absorbance of 0.7 (±0.02) at λ = 735 nm. The reaction was started by adding 300 μL of prewarmed (30 °C) $ABTS^{\bullet+}$ solution. The absorbance was measured at λ = 735 nm exactly after incubation for 30 min at 30 °C using a microplate reader (TECAN safire^2, Crailsheim, Germany). The microplate was shaken prior to reading. The extracted apple samples were dissolved in phosphate buffer to fit in the concentration range of the Trolox calibration curve. All reaction mixtures were prepared in triplicate.

The TE concentrations of the apples were calculated by subtracting the blank from samples and standards using a linear calibration curve constructed for each assay. One TE equals the net protection produced by 1 mM Trolox.

Statistical Analysis. All statistical calculations were performed using the STATVIEW program version 5.0 (SAS Institute, Cary, NC; 1998). Results were reported as mean ± standard deviation (SD). Differences between the mean values of polyphenols and antioxidant capacity in the different agricultural systems were statistically analyzed using a three-way analysis of variance (ANOVA) including year, farm pair (site), production method, and corresponding interactions and the Tukey−Kramer post hoc test. Year-to-year differences were statistically analyzed using repeated-measures ANOVA and the Tukey−Kramer post hoc test. Polyphenol concentrations were transformed logarithmically because the equal variance and normal assumption of ANOVA were rejected. The relationship between the polyphenol content and the antioxidant capacity in apples was analyzed by applying stepwise iterated multiple linear regression modeling. Differences were considered to be significant at $p < 0.05$.

RESULTS

The antioxidant status and the polyphenol concentrations were analyzed in five comparable farm pairs (FP; organic, conventional) for a period of three years (2004−2006). Additionally, the year-to-year variations were calculated. Finally, we analyzed the single and multiple correlations between the concentration of the different polyphenol compounds and the antioxidant capacity.

Quality Parameters. The fruit weight of the apples ranged from 146.7 to 197.8 g. Throughout the study significant differences

Table 3. Concentrations of Phenolic Compounds in Apples (Cv. 'Golden Delicious') from Organic (org) and Conventional (conv) Production Methods in 2004 (**A**), 2005 (**B**), and 2006 (**C**)[a]

		chlorogenic acid (μg/g of FW)	hydroxycinnamic acids (μg/g of FW)	flavanols (μg/g of FW)	dihydrochalcones (μg/g of FW)	flavonols (μg/g of FW)	sum of phenolic compounds (μg/g of FW)
				(**A**) 2004			
FP1	org	104.2 ± 25.5	28.3 ± 5.3	120.8 ± 30.5	25.3 ± 5.2	29.2 ± 10.5	307.8 ± 70.9
	conv	96.7 ± 10.8	31.6 ± 4.2	121.2 ± 24.6	24.3 ± 5.1	46.8 ± 27.7	320.6 ± 64.3
FP2	org	84.9 ± 25.6 a	26.4 ± 3.5	123.8 ± 28.0	23.4 ± 5.2 a	57.7 ± 29.4 a	316.2 ± 73.7
	conv	72.4 ± 17.4	23.5 ± 3.2	152.7 ± 38.9	20.5 ± 3.6	38.8 ± 17.5	307.8 ± 60.3
FP3	org	100.8 ± 14.6 a	26.3 ± 4.7	167.3 ± 41.0	25.7 ± 4.5 a	43.0 ± 5.9	363.2 ± 57.1 a
	conv	78.7 ± 15.9	25.8 ± 4.5	113.9 ± 39.4	17.9 ± 4.2	46.0 ± 14.3	282.4 ± 67.9
FP4	org	86.3 ± 11.3 a	24.2 ± 3.8	65.5 ± 15.9	21.9 ± 3.7 a	63.9 ± 15.3 a	261.8 ± 35.9 a
	conv	76.5 ± 13.7	24.1 ± 2.3	64.1 ± 17.9	14.9 ± 1.4	39.3 ± 12.9	219.0 ± 25.0
FP5	org	58.6 ± 14.8	19.6 ± 3.1	118.0 ± 23.9	19.4 ± 5.5	46.9 ± 14.5 a	262.5 ± 50.8
	conv	90.6 ± 22.2 b	27.8 ± 2.9 b	102.8 ± 56.1	20.9 ± 3.4	29.6 ± 19.3	271.7 ± 81.7
mean	org	86.9 ± 25.0	24.9 ± 5.0	121.9 ± 41.8	23.2 ± 5.3	47.3 ± 20.1	304.4 ± 68.6
	conv	83.3 ± 18.3	26.7 ± 4.5	113.4 ± 45.8	19.9 ± 4.7	40.1 ± 19.4	283.6 ± 69.4
				(**B**) 2005			
FP1	org	83.3 ± 13.2	18.2 ± 3.2	89.5 ± 8.5	26.4 ± 3.7	53.4 ± 18.9	270.8 ± 39.5
	conv	95.8 ± 8.9	18.7 ± 3.7	106.9 ± 12.4 b	29.9 ± 1.9	56.8 ± 12.5	308.1 ± 23.8
FP2	org	73.2 ± 14.4	16.8 ± 3.7 a	84.1 ± 10.9 a	19.3 ± 7.2	69.5 ± 25.3	262.8 ± 55.9
	conv	73.7 ± 12.9	13.9 ± 2.2	78.5 ± 13.7	22.2 ± 4.0	60.1 ± 25.9	248.5 ± 34.5
FP3	org	87.1 ± 8.1 a	24.0 ± 2.6 a	99.4 ± 8.7 a	23.9 ± 2.8 a	62.2 ± 32.1	296.6 ± 41.8 a
	conv	58.6 ± 6.8	16.2 ± 0.6	80.8 ± 12.2	20.4 ± 2.7	55.1 ± 28.5	231.1 ± 27.9
FP4	org	96.6 ± 21.4 a	18.7 ± 5.4 a	108.9 ± 12.5 a	32.5 ± 9.5 a	76.8 ± 30.0	333.6 ± 70.0 a
	conv	65.2 ± 11.2	12.8 ± 2.0	87.9 ± 17.3	21.6 ± 2.2	59.7 ± 10.2	247.4 ± 27.9
FP5	org	77.8 ± 5.5 a	18.1 ± 1.7 a	109.1 ± 5.8 a	23.6 ± 2.6 a	116.9 ± 45.7	345.5 ± 50.4 a
	conv	48.5 ± 10.7	12.9 ± 1.1	92.3 ± 13.3	19.3 ± 3.7	58.3 ± 18.0	231.4 ± 42.8
mean	org	83.6 ± 14.9 a	19.1 ± 4.1 a	98.2 ± 13.5 a	25.2 ± 6.9	75.8 ± 36.6	301.9 ± 58.7 a
	conv	68.4 ± 18.8	14.9 ± 3.0	89.3 ± 16.3	22.6 ± 4.7	58.0 ± 18.7	253.3 ± 41.3
				(**C**) 2006			
FP1	org	120.6 ± 11.5 a	26.6 ± 2.9 a	234.7 ± 38.6 a	38.6 ± 7.4 a	74.9 ± 31.8	495.5 ± 77.7 a
	conv	105.4 ± 8.4	21.8 ± 3.5	148.4 ± 40.1	33.6 ± 2.6	54.0 ± 10.4	363.3 ± 39.0
FP2	org	110.6 ± 2.0 a	45.7 ± 14.1 a	266.6 ± 62.3 a	30.6 ± 4.3 a	47.1 ± 20.0	500.6 ± 64.9 a
	conv	91.9 ± 14.7	19.6 ± 3.5	173.6 ± 19.5	22.2 ± 4.1	34.2 ± 12.8	341.5 ± 41.4
FP3	org	79.8 ± 6.7 a	22.1 ± 2.5	153.5 ± 5.3	27.7 ± 2.5 a	49.0 ± 14.8	332.2 ± 23.1
	conv	72.7 ± 6.6	22.7 ± 4.5	175.7 ± 33.9 b	25.4 ± 2.9	32.1 ± 11.3	328.7 ± 42.1
FP6	org	108.9 ± 14.0	29.9 ± 6.8	157.7 ± 46.2	31.4 ± 3.9	48.3 ± 19.2	376.4 ± 80.9
	conv	116.7 ± 13.2 b	29.3 ± 2.3	179.3 ± 18.3 b	32.1 ± 4.8	49.3 ± 23.5	406.8 ± 47.3
FP7	org	76.7 ± 8.2	26.8 ± 7.9	147.5 ± 16.7	25.5 ± 3.1	30.3 ± 11.5	306.9 ± 39.5
	conv	94.8 ± 7.9 b	27.1 ± 5.1	199.9 ± 61.0 b	27.2 ± 1.5	33.7 ± 14.1	382.8 ± 84.5
mean	org	99.3 ± 20.0	30.3 ± 11.1 a	192.0 ± 61.7	30.7 ± 6.2	49.9 ± 23.8	402.3 ± 100.2
	conv	96.3 ± 17.8	24.1 ± 5.1	175.4 ± 38.5	28.1 ± 5.3	40.6 ± 16.7	364.6 ± 56.7

[a] Apples were harvested from five comparable farms pairs (FP) in Switzerland. Values are means ± SD; $n = 5$ per group (a = organic significant higher than conventional ($p < 0.05$); b = conventional significantly higher than organic ($p < 0.05$); three-way ANOVA; Tukey−Kramer post hoc test). The "sum of phenolic compounds" is calculated as the sum of the results for the individual polyphenols: chlorogenic acid, hydroxycinnamic acids (4-caffeoylquinic acid, 3-coumaroylquinic acid, 4-coumaroylquinic acid, 5-coumaroylquinic acid), dihydrochalcones (phloretin 2′-xyloglucoside, phloretin 2′-glucoside), flavanols (catechin, epicatechin, procyanidin B1, procyanidin B2), and flavonols (quercetin 3-galactoside, quercetin 3-glucoside, quercetin 3-xyloside, quercetin 3-arabinoside, quercetin 3-rhamnoside).

were observed in the organically (163.7 ± 13.5 g) and conventionally (178.7 ± 13.5 g) produced apples.

No significant differences (organic vs conventional) were found in the parameters fruit flesh firmness (5.5 ± 1.4 kg/cm^2 organic vs 5.6 ± 1.4 kg/cm^2 conventional) and sugar content (12.9 ± 1.5 °Brix organic vs 12.9 ± 1.0 °Brix conventional). Additionally, the significantly highest sugar contents (2004, 12.1 ± 0.6 °Brix; 2005, 12.9 ± 0.9 °Brix; 2006, 14.2 ± 1.3 °Brix) were observed in 2006.

Dry Matter. The dry matter of the analyzed apples ranged from 10.6 to 29.4%. No significant differences were observed between the organically (16.0 ± 3.1%) and conventionally produced apples (16.9 ± 3.9%) throughout the years (data not shown). Therefore, in the following the results are expressed in fresh weight (FW).

Antioxidant Capacity. The antioxidant capacity of the apple extracts was measured using three different antioxidant assays (FRAP, TEAC, and ORAC assays). The results are listed in **Table 2**.

In 2004, there were no significant differences in the antioxidant capacities between the five FPs using the ORAC assay. However, using the FRAP and TEAC assays, organically produced apples showed a 7−27% significantly higher antioxidant capacity than the conventionally produced apples (**Table 2A**) in three FPs.

Overall, in 2005 and 2006, the organically produced apples revealed a significantly higher (15%, $p < 0.05$) antioxidant capacity than the conventionally produced fruits (**Table 2B**,**C**).

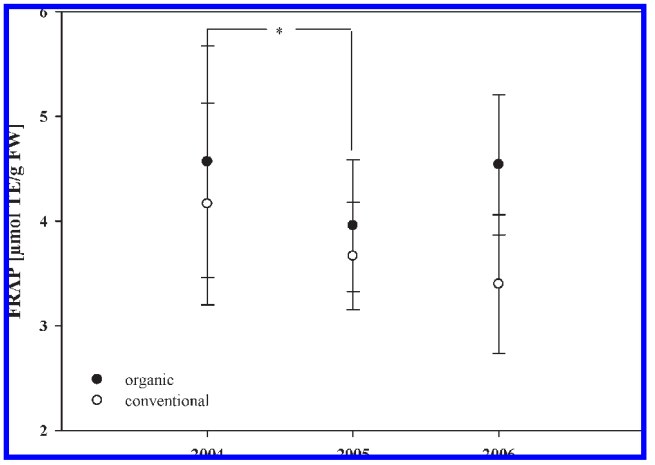

Figure 2. Changes in antioxidant capacity over three years (2004−2006) in three farm pairs. Significant differences between crop years 2004 and 2005 (∗, $p < 0.05$, repeated measures ANOVA; Tukey−Kramer post hoc test) were observed. TE, Trolox equivalent; FW, fresh weight.

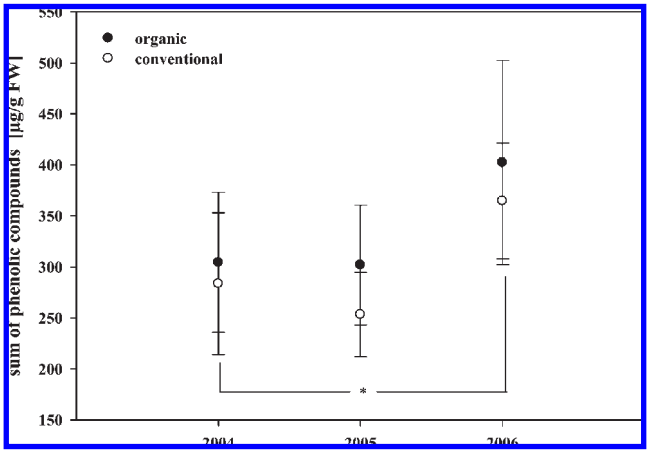

Figure 3. Phytochemical concentration over three years (2004−2006) in three farm pairs. Significant differences between crop years 2004, 2005, and 2006 (∗, $p < 0.05$, repeated measures ANOVA; Tukey−Kramer post hoc test) were observed. The "sum of phenolic compounds" is calculated as the sum of the results for the individual polyphenols: chlorogenic acid, hydroxycinnamic acids (1,4-caffeoylquinic acid, 3-coumaroylquinic acid, 4-coumaroylquinic acid, 5-coumaroylquinic acid), dihydrochalcones (phloretin 2′-xyloglucoside, phloretin 2′-glucoside), flavanols (catechin, epicatechin, procyanidin B1, procyanidin B2), and flavonols (quercetin 3-galactoside, quercetin 3-glucoside, quercetin 3-xyloside, quercetin 3-arabinoside, quercetin 3-rhamnoside).

Concentration of Phenolic Compounds. The polyphenol classes identified in apples using HPLC-MS were hydroxycinnamic acids (chlorogenic acid, 4-caffeoylquinic acid, 3-coumaroylquinic acid, 4-coumaroylquinic acid, 5-coumaroylquinic acid), dihydrochalcones (phloretin 2′-xyloglucoside, phloretin 2′-glucoside), flavanols (catechin, epicatechin, procyanidin B1, procyanidin B2), and flavonols (quercetin 3-galactoside, quercetin 3-glucoside, quercetin 3-xyloside, quercetin 3-arabinoside, quercetin 3-rhamnoside). Because chlorogenic acid is the predominant polyphenol compound found in apple extracts, it is listed separately. Otherwise, the polyphenol classes are listed as the sum of the single compounds. **Figure 1** shows representative HPLC chromatograms.

In 2004, significant differences were observed between chlorogenic acid ($p = 0.009$), flavonols ($p = 0.004$), and dihydrochalcones ($p < 0.001$) in three of five FPs and also between the flavanols ($p = 0.03$) in one of five FPs. The organically produced apples showed higher concentrations of chlorogenic acid, flavonols, flavanols, and dihydrochalcones than the conventionally produced fruits (**Table 3A**).

In 2005, the hydroxycinnamic acid content was significantly higher in the apples from organic orchards in four FPs. Furthermore, in 2005 the flavanol concentration was higher in four FPs with organic production and the following year in two FPs.

In 2005 and 2006, no statistically significant influences of the production method were observed concerning the contents of flavonols. However, the dihydrochalcone and chlorogenic acid concentrations were higher in organically produced apples in three of five FPs, as listed in **Table 3B**,**C**.

Influence of the Cropping Year. Three of five FPs were identical over the three cropping years (FPs 1−3, **Tables 2** and **3**). Therefore, year-to-year variations of the polyphenol content as well as the antioxidant capacity were only analyzed in these three FPs by applying a three-way ANOVA (location, production method, year).

In 2004, significantly higher antioxidant capacities (FRAP, ORAC, and TEAC assays) were found than in 2005 (**Table 2A**,**B**). No further significant differences were observed between the other crop years (2005 and 2006). As an example, the results of the FRAP assay are shown in **Figure 2**.

In 2005, the lowest hydroxycinnamic acid concentrations (including chlorogenic acid) were observed. No significant year differences were found between the other harvest years. In 2006, the dihydrochalcones and flavanols exhibited the highest concentrations with proportions of 8.6 and 54.0% of the total polyphenol concentration. The highest flavonol concentrations were found in 2005, reaching 66.9 µg/g of FW. When the sums of phenolic compounds were compared, the highest concentrations were detected in 2006 independent of the production method (**Figure 3**).

In all three years, the levels of hydroxycinnamic acids, dihydrochalcones, and flavanols and also the antioxidant capacity were statistically higher in organically produced apples in the magnitude of 14−19%.

Multiple Regression of the Antioxidant Capacity of Different Polyphenol Components. A multiple linear regression analysis of the results from all analyzed apples was performed to predict the antioxidant capacity versus the concentration of specific polyphenolic compounds. Significantly positive correlations were observed between the antioxidant capacity of apples and their polyphenol contents (**Table 4**).

DISCUSSION

Few data exist regarding the influence of the production method (organic vs conventional) on the phytochemical concentration in apples sampled for several years. Therefore, the aim of the present study was to evaluate the phytochemical concentration and the antioxidant capacity of apples grown under well-defined organic and conventional conditions for 3 years.

The phytochemical concentration, antioxidant capacity, and dry matter of the apples (cv. 'Golden Delicious') were similar to those previously reported (*20*, *24*, *25*). The antioxidant capacity of the apple extracts can be attributed to the phytochemical content because the phytochemical concentration was positively correlated with the antioxidant capacity of fruits including apples (*20*, *26*, *27*). The association between the antioxidant capacity and the polyphenol content of our study is well in line with those previously reported (*20*, *28*, *29*). However, the different phenolic classes correlated differently with the antioxidative capacity. In the present study, the FRAP assay is primarily determined by the procyanidin content (flavanol), whereas the TEAC assay depends

Table 4. Multiple Regression Analysis of Apples (Cv. 'Golden Delicious') with the Antioxidant Capacity as Dependent Variable[a]

P values						
chlorogenic acid	dihydrochalcones	flavanols	flavonols	hydroxycinnamic acids	R^2 adj	antioxidant assay
<0.001		<0.05	<0.001	<0.001	**0.596**	FRAP
		<0.001	<0.001	<0.0001	**0.432**	ORAC
<0.0001	<0.01		<0.0001		**0.495**	TEAC

[a] Multiple regression analysis involving chlorogenic acid, dihydrochalcones (phloretin 2′-xyloglucoside, phloretin 2′-glucoside), flavanols (catechin, epicatechin, procyanidin B1, procyanidin B2), flavonols (quercetin 3-galactoside, quercetin 3-glucoside, quercetin 3-xyloside, quercetin 3-arabinoside, quercetin 3-rhamnoside), hydroxycinnamic acids (4-caffeoylquinic acid, 3-coumaroylquinic acid, 4-coumaroylquinic acid, 5-coumaroylquinic acid) as independent variables was performed. Only significant variables are shown.

more on the content of quercetin glycosides (flavonol). This is in agreement with other studies (*28*, *29*). In the present study the flavonols and flavanols had a large influence on the antioxidant capacity. To date, no study has been published reporting the correlations between the ORAC assay and polyphenol concentrations. We were able to show that the ORAC assay was primarily determined by the flavanols. We, therefore, suggest that flavanols and flavonols are the strongest antioxidants in apples. Due to their structural characteristics they are potent antioxidants. The *o*-dihydroxy moiety in the B-ring and the *cis*-hydroxyl group in the C-3 of the C-ring stabilize the resulting free radical form (*29*, *30*).

The organically grown apples showed a tendency of higher phytochemical concentrations compared to the conventionally produced apples (10%), resulting in a 12% higher antioxidant capacity in the course of 3 years. This is in agreement with the studies of Weibel et al. (*16*, *17*). The authors reported a polyphenol concentration up to 23% higher in organically produced apples. In other studies polyphenol concentration and antioxidant capacity were significantly higher in apples from integrated production (*18*, *20*) or equal (*18*, *19*) to those of organically produced ones. However, these studies were conducted using fruits of only one harvest as well as different cultivars. To date, only three studies analyzed the polyphenol concentrations in tomatoes and apples over a period of several years (*17*, *31*, *32*). In both tomato studies the flavonoid concentration in organically produced fruits was higher than in the conventionally produced ones. Furthermore, the phytochemical concentration in the organically produced apples was higher (*17*). The tendency of higher polyphenol concentrations in organically produced fruits and vegetables could be explained by a higher phosphorus uptake and limited nitrogen availability (*9*, *16*, *33*). An increased phosphorus uptake can provide the necessary energy for the synthesis of phytochemicals (*16*). Furthermore, it has been shown that plants synthesized more flavonoids when nitrogen was limited (*32*, *34*).

In this study significant but not large differences were observed in the phytochemical content and the antioxidative capacity between organically and conventionally produced apples in some cases. An explanation for the rather small differences could be that cultivar and climate, which were the same in both groups, are key determinants for the concentration of phytochemicals in apples. In addition, year-to-year variations (up to 20%) in both groups were larger than the differences between the agricultural systems within 1 year. Factors such as plant genotype, cultivar, and climate variations have been shown to have a great influence on the phytochemical content (*9*, *35*−*37*), confirming our observations. The differences in the crop years could be explained by climate variations. In 2006 20% less rainfall was observed than in 2005 (84.7 L/m^2), which may explain the higher polyphenol concentration in 2006. One might speculate that more sunshine resulted in the significantly higher sugar contents (°Brix) of apples in harvest year 2006. Photosynthetic performance is important for the synthesis of polyphenols. Another parameter, that is, temperature, was comparable throughout the three years (average temperature = 15 °C during the vegetation period). However, the antioxidant capacity and the phytochemical concentration did not differ to the same extent from year to year. It might be speculated that other antioxidant substances were influenced by climate factors as well.

In conclusion, in the present study the organically produced apples displayed a higher phytochemical concentration and a higher antioxidant capacity than conventionally produced apples. However, it remains unclear whether these minor differences caused by the production method are of nutritional relevance.

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