Changes in Tea Performance and Soil Properties after Three Years of Polyhalite Application

Zijun Zhou, Kun Chen, Hua Yu, Qingrui Chen,* Fachi Wu,
Xiangzhong Zeng, Shihua Tu, Yusheng Qin,* Robert Meakin, and Xiaohui Fan

ABSTRACT

Tea (*Camellia sinensis* L.) is the world's most widely consumed beverage with high economic and health benefits. Potassium is the second key nutrient for tea plants. Polyhalite is applied in some grain crops as a novel source of K, which also contains Ca, Mg, and S. However, the feasibility of using polyhalite for tea growth is unclear. Consequently, a 3-yr field experiment was conducted to determine effects of polyhalite on soil properties, tea yield, economic benefits, and tea quality in southwest China. There were three treatments: control (CK, without K fertilizer), sulfate of potash (SOP) and polyhalite (POLY4). Compared with CK, SOP and POLY4 application increased soil available K, P, and S contents, as well as tea yields and economic benefits. Compared with SOP, POLY4 increased soil exchangeable Ca and Mg and available S contents, but decreased soil acidification due to high addition of $CaSO_4$ and $MgSO_4$. POLY4 application led to 15.1% higher tea yield than SOP in 2017, while no difference was observed in 2015 and 2016. The POLY4 was economically superior to SOP in 2017 with a greater net benefit by US\$1982 ha^{-1}. Although the two treatments did not differ on tea quality, correlation analysis demonstrated that quality was generally positively and significantly correlated with K, Ca, Mg, P, and S contents in tea leaves. Consequently, POLY4 was suitable as a K source for tea. Given high S and Ca contents in POLY4, it might be better to apply POLY4 together with other K fertilizers in future studies.

Core Ideas

- Application of polyhalite increased tea yield and economic benefits.
- Polyhalite application showed no adverse effects on tea quality.
- Soil acidification was significantly reduced with polyhalite.
- Polyhalite would be widely used as a novel potassium fertilizer in tea plantations.

Tea is one of the world's most popular and widely consumed beverages and is beneficial to human health because it contains antioxidants and has anti-aging effects (Li et al., 2011a; Fei et al., 2017). In 2016, global tea production was 5.95 million tons from 4.10 million hectares. China was the largest tea exporter, and in 2016 accounted for 40.5% of global production (FAOSTAT, http://faostat.fao.org). Tea is also one of the major cash crops in tropical and subtropical China (Ruan et al., 2013), especially in Sichuan, Yunnan, and Guizhou provinces in south-western China, which is a major tea-growing region of China (Su et al., 2017). The region is characterized by high rainfall and acidic soils with low fertility. Since these soils are low in basic cations, such as K, Ca, and Mg, that are required for tea growth, fertilization is necessary during tea plantation (McKenzie et al., 2004; Alekseeva et al., 2011; Li et al., 2016).

Potassium is the second key nutrient after N for tea growth (Rajan and Anandhan, 2016; Singh and Pathak, 2018). The positive response of tea in terms of both yield and quality to K application has been reported in many studies, especially in soils containing small amounts of exchangeable K (Venkatesan et al., 2005; Ruan et al., 2013). Although there are two commonly available forms of K-fertilizer (SOP and MOP) (Singh and Pathak, 2018), the responses of tea yield and quality to MOP have been variable (Ruan et al., 1998, 2007, 2013; Venkatesan et al., 2005). Ruan et al. (1998) demonstrated that MOP had detrimental effects on accumulation of free amino acids in young tea plants in a plot experiment, but found comparable effects of SOP and MOP on total free amino acids in mature tea plants under field conditions (Ruan et al., 2013). Similar detrimental effects of Cl from MOP on N uptake and concentration of theanine in young shoots was observed by Ruan et al. (2007). Venkatesan et al. (2005) found that, compared to MOP, a comparatively small quantity of SOP could improve the same parameters of tea. In addition, soil S availability was found to be important in optimizing tea production (Karak et al., 2015). Therefore, SOP is a priority K fertilizer applied in many tea plantations in China (Yang et al., 2018).

Canada, Russia, and Belarus abound in K-rich minerals and produce more than 90% of the world's potash (Shekhar et al.,

Published in Agron. J. 111:1967–1976 (2019)
doi:10.2134/agronj2018.06.0393
Supplemental material available online

Z. Zhou, K. Chen, H. Yu, Q. Chen, F. Wu, X. Zeng, S. Tu, Y. Qin, Soil and Fertilizer Institute, Sichuan Academy of Agricultural Sciences, Chengdu 610066, China; Z. Zhou, K. Chen, H. Yu, Q. Chen, F. Wu, X. Zeng, S. Tu, Y. Qin, Monitoring and Experimental Station of Plant Nutrition and Agro-Environment for Sloping Land in South Region, Ministry of Agriculture and Rural Affairs, Chengdu 610066, China; R. Meakin, X. Fan, Sirius Minerals Plc, Scarborough YO11 3ZB, United Kingdom. Received 20 June 2018. Accepted 5 Mar. 2019. *Corresponding authors (2337504329@qq.com; shengyuq@126.com).

Abbreviations: MOP, muriate of potash; POLY4, polyhalite; SOP, sulfate of potash.

2017). However, China is deficient in K resources and meets most of its requirements of K fertilizers through imports, and the expensive imported fertilizers reduce economic benefits to farmers. Some K-bearing minerals and rocks, such as crushed biotite and vermiculite, can be used as fertilizers to alleviate K deficiency in soil and to reduce production costs (Li et al., 2015). Polyhalite ($K_2SO_4{\cdot}MgSO_4{\cdot}2CaSO_4{\cdot}2H_2O$) is a marine evaporite mineral produced by successive marine evaporation events throughout history (Kemp et al., 2016), and shows huge potential as K fertilizer due to its very low quantities of Cl but rich in K, Ca, and Mg (Ogorodova et al., 2016; Albadarin et al., 2017). In addition to K, Mg fertilization can increase tea yield and quality (Jayaganesh et al., 2006; Ruan et al., 2012). Calcium is also a necessary nutrient for tea plants and can increase tea growth after application, although some research proposed that tea is a calcifuge and acidophilic plant (Karak et al., 2017), and excess Ca in soil can inhibit K uptake by tea (Fung and Wong, 2004). Some studies have examined polyhalite effects on crops. Polyhalite was as effective as SOP for maize (*Zea mays* L.) and sorghum (*Sorghum bicolor* L. Moench) in Fraps and Schmidt (1932), and for potato (*Solanum tuberosum* L.) and flax *(Linum usitatissimum* L.) production by Lepeshkov and Shaposhnikova (1958). Panitkin (1967) demonstrated that polyhalite performed better than SOP in potato and beet (*Beta vulgaris* L.) because of Mg supplied by polyhalite. Compared to soluble sulfate sources of K, Mg, and Ca, Barbarick (1991) found that polyhalite produced higher total yields of sorghum-sudangrass (*Sorghum bicolor* L. Moench, *Sorghum sudanense* Piper Stapf). However, the effects of applying polyhalite in tea plantations as a source of K have received little attention.

The extraction and use of polyhalite is difficult and limited because of its complex composition, weak solubility, deep burial (below 1000 m), and the relatively minor deposits in some countries, such as China (Zhao et al., 2011). Recently, North Yorkshire, UK, was reported to hold the largest and highest-quality deposits of polyhalite (2660 Mt of 85.7% purity) in the world, and could be mined and marketed with no processing except crushing and sizing (Kemp et al., 2016). We hypothesize that polyhalite fertilizer may be well suited to tea plantations, because it is rich in K, Ca, and Mg and tea soil is often deficient in these basic cations (Fung and Wong, 2004; Yang et al., 2018). The aim of the study was to examine the feasibility of polyhalite as a K fertilizer in tea plantations by determining: (i) changes in physicochemical properties of soil; (ii) tea quality; and (iii) tea yield and economic benefits.

MATERIALS AND METHODS

Experimental Site and Materials

Field trials were conducted for three consecutive years, from June 2014 to August 2017 in E'mei county (29°40′27″ N, 103°30′51″ E, elevation 450 m), Sichuan province, China. The mean annual temperature is 18.2°C and the mean annual precipitation is 1555.3 mm. The soil, classified as a Hapludult according to the US soil taxonomy, is a loamy clay with 34% sand, 26.4% silt, and 39.6% clay. Before the experiment, the basic physicochemical properties of the surface soil (0–20 cm) were as follows: pH (extract of soil in water), 4.46; organic matter, 14.25 g kg^{-1}; available N, 101.60 mg kg^{-1}; available P, 6.50 mg kg^{-1}; available K, 60.00 mg kg^{-1}; exchangeable Ca, 1.60 g kg^{-1}; exchangeable Mg 88.71 mg kg^{-1}; and available S, 126.45 mg kg^{-1} (Lu, 1999; Lu et al., 2014).

The variety of tea was Wuniuzao, which is one of the most widespread varieties locally due to its distinctive elite features of quality and yield, as well as early break property of young shoots in spring (Li et al., 2011b; Zhao et al., 2016). Before the experiment, the tea plants grew for 14 yr in the studied tea plantation. The polyhalite fertilizer chosen for the experiment was granular POLY4, supplied by Sirius Minerals Plc, Scarborough, UK. It was made of polyhalite powder, with specified composition as follows: 14% K_2O, 6% MgO, 17% CaO, and 19% S. The SOP used for comparison, supplied by K+S Group, Kassel, Germany, contained 50% K_2O and 17% S. The N fertilizer was applied in the form of commercial-grade urea (460 g N kg^{-1}) and P fertilizer, as mono-ammonium phosphate (610 g P_2O_5 kg^{-1}, 100 g N kg^{-1}), both supplied by Xinlianxin Co. Ltd., Henan, China.

Experimental Treatments

The experiment was laid out in a randomized design with three replications. Each plot measured 20 m^2 (13.33 m by 1.5 m) and consisted of one 13.33-m-long line of tea bush. Each tea bush was planted in double rows with inner row distance of 0.3 m and outer row distance was 1.2 m. A buffer zone of 0.15 m separated adjacent plots, making the distance between tea rows in adjacent plots at least 1.35 m. The experiment comprised three treatments, namely a control (CK, without any K fertilizer), SOP at 180 kg K_2O ha^{-1}, and polyhalite (POLY4) at 180 kg K_2O ha^{-1}. The annual dose of N (240 kg N ha^{-1}) and of P (120 kg P_2O_5 ha^{-1}) was common to all the treatments including CK. Nitrogen and K were applied by splitting the total dose into two equal doses, a basal dose around 15 November along with all of P at the dormant phase of the aboveground tea, and as a top dressing around 12 February (~10 d before spring tea picking). Fertilizers were applied by hand as band application in the inter-row of tea bushes in each plot. The time of tea pruning was around 24 March between spring and summer in each year during the experiment (Han et al., 2007).

Physicochemical Properties of Soil

After the summer harvest each year, four soil cores (5 cm diameter, 20 cm length) from the 0- to 20-cm plow layer were randomly collected between the double rows (~15 to 20 cm from tea plants) of tea bush from each replication and pooled to make one composite sample for each plot. After removing visible stones and plant residues, the soil was air-dried in the shade at room temperature for approximately 30 d, and then homogenized, and passed through a 2-mm mesh. Some basic physicochemical properties of the soil were analyzed, namely the contents of available N, P, K, Mg, Ca, and S, and soil pH. Available N was determined by the alkali solution diffusion method; available P, extracted by 0.025 M HCl–0.03 M NH_4F and determined by ammonium molybdate colorimetry; and available K, extracted by 2 M HNO_3 and determined by atomic absorption photometry (Lu, 1999). The homogenized soil was suspended in 0.1 M $BaCl_2$ (1:50, w/v) for 30 min; the suspension was passed through a 0.45-mm filter, and exchangeable Ca and Mg contents of the filtrate were determined using inductively coupled plasma atomic emission spectroscopy (ICP–AES) (Lu et al., 2014). Available S in the soil was extracted by shaking 2.5 g of soil with 25 mL of Mehlich-3 solution for 5 min and determined using ICP–AES (Yang et al., 2018). Soil pH (1:2.5

Table 1. Effects of source of K on soil pH, available N (N_A), available P (P_A), available K (K_A), exchangeable Ca (Ca^{2+}_{E}), exchangeable Mg (Mg^{2+}_{E}), and available S (S_A).

Year	Treatment†	pH	N_A	P_A	K_A	Ca^{2+}_{E}	Mg^{2+}_{E}	S_A
			mg kg^{-1}					
2015	Control	4.22 ± 0.08ab‡	113.8 ± 10.4a	7.12 ± 0.91a	68.7 ± 6.4a	1609 ± 61a	77.5 ± 6.2a	128.6 ± 2.8a
	SOP	4.05 ± 0.16a	123.3 ± 4.7a	7.65 ± 1.05a	109.3 ± 4.2b	1598 ± 63a	89.1 ± 12.6a	137.3 ± 4.7b
	POLY4	4.43 ± 0.07b	120.0 ± 3.3a	7.69 ± 1.95a	101.7 ± 1.52b	1736 ± 108a	121.2 ± 7.6b	143.1 ± 5.0b
2016	Control	4.03 ± 0.06b	105.4 ± 13.5a	9.16 ± 0.20a	75.7 ± 3.1a	1522 ± 52a	82.4 ± 11.2a	141.4 ± 4.2a
	SOP	3.80 ± 0.10a	119.3 ± 15.0a	13.13 ± 1.32b	123.7 ± 4.2b	1590 ± 52a	80.2 ± 3.2a	155.4 ± 5.1b
	POLY4	4.29 ± 0.11c	115.2 ± 15.3a	12.37 ± 0.69b	127.7 ± 3.8b	2044 ± 170b	128.8 ± 11.1b	197.9 ± 6.8c
2017	Control	3.91 ± 0.02b	104.3 ± 6.9a	10.41 ± 0.25a	77.7 ± 2.5a	1540 ± 160a	81.6 ± 5.4a	149.2 ± 3.4a
	SOP	3.72 ± 0.02a	123.7 ± 11.3a	12.77 ± 1.05b	179.7 ± 5.0b	1624 ± 115a	84.2 ± 8.6a	190.2 ± 8.2b
	POLY4	3.97 ± 0.03c	110.3 ± 6.5a	12.46 ± 1.70b	170.0 ± 27.8b	2196 ± 173b	134.4 ± 3.9b	239.3 ± 10.2c

† SOP, commercial sulfate of potash; POLY4, polyhalite.

‡ Values are mean ± standard deviation (*n* = 3). Means with the same letters in the same column for the same year are not significantly different by Tukey test ($P > 0.05$).

soil/water) was measured using an Orion 3-star benchtop pH meter (Thermo Scientific, Waltham, MA).

Total N, P, K, Ca, Mg, and S in Tea Leaves

Green tea was processed by the modified method described by Lin et al. (2012): freshly harvested shoots were kept under shade for 5 h and microwaved for 80 s to inactivate the enzymes in a microwave oven (Galanz B8023CSL-K3, Guangdong, China). The shoots were then rolled by hand for 10 min and the rolled shoots dried for 10 h at 80°C in an electrically-heated drum wind drying oven (Keelrein Instrument Co. Ltd., Shanghai, China). Of dried subsamples, 100 g was ground to pass through a 40-mesh sieve (particle size 0.42 mm). The ground tea leaves (1 g) were digested in a mixture of concentrated nitric acid (HNO_3) and concentrated perchloric acid ($HClO_4$; 5:1, v/v) (Lu, 1999). Total N was measured using a Model 1500 CNS Analyzer (Carlo Erba Strumentazione, Milan, Italy); P, K, Ca, Mg, and S by ICP–AES using an IRIS/AP plasma spectrometer (Thermo Jarrell Ash, Mount Holly, NJ).

Assessment of Tea Quality

Total amino acids in tea leaves were determined according to Chinese National Standard GB/T 5009.124 (2016) and Xu et al. (2017). One gram of tea powder was added to a glass tube containing 6 M HCl solution and then exposed to blown nitrogen gas for 5 min. After that, the mixture was hydrolyzed for 22 h at 110°C. The hydrolysate was centrifuged to remove any sediment and the supernatant evaporated using a rotary evaporator at 50°C. The residue was dissolved in 2 mL of sodium citrate buffer solution (pH 2.2) and filtered through a 0.22-mm membrane, after which the filtrate was then analyzed using an automatic amino acid analyzer. Total polyphenols in tea leaves were measured by the Folin–Ciocalteu method according to Chinese National Standard GB/T 8313 (2008) and Zeng et al. (2017). Finely powdered sample (200 mg) was extracted twice with 4 mL of 70% methanol solution for 10 min at 75°C. The extracts were combined and diluted to 10 mL with the extraction solvent. The solution was filtered through a 0.45-µm membrane extract and diluted 100-fold with distilled water. Diluted extract or standard gallic acid solution (1 mL) was decanted into a 15-mL centrifuge tube. Then 5 mL of 10% Folin–Ciocalteu reagent was added to the tube and the mixture shaken. After 4 min, 4 mL of 7.5% Na_2CO_3 solution was added to the mixture. After incubating for 60 min at room temperature, the absorbance was measured at 765 nm to measure total polyphenols. According to Chinese National Standard GB/T 8305 (2013), mixtures of 2.0 g of tea powder were placed in 300 mL of boiling water and maintained in a water bath for 45 min at 100°C and then filtered. The filtered residue was used to calculate the water extract contents after oven drying at 120°C to constant weight. The ratio of polyphenols to total amino acids was also calculated.

Tea Yield

Tea was harvested twice a year by hand in the studied area: spring tea (around 20 February to 15 March) and summer tea (around 1 to 30 April). The spring harvest comprised four pickings in 2015 and 2017 and three pickings in 2016, which was a low-rainfall year, and the mean temperature during picking of spring tea was around 11°C. The pickings were in the form of a bud and two adjacent expanding leaves. The summer harvest comprised three pickings, which were limited to a bud and only one leaf, and the mean temperature during picking of summer tea was around 21°C. Around a week elapsed between two adjacent picking times. The tea was harvested in each total plot and the yield measured. The total fresh yield was the sum of the two harvests.

Statistical Analysis

All the values were compared using one-way analysis of variance (ANOVA) with Tukey test at a probability level of 0.05 on SPSS ver. 16.0 (IBM Corp., Armonk, NY). Prior to analysis, the data were tested for homogeneity of variance using Levene's test. Pearson's correlation analysis was used to investigate the relationships among tea yield, soil physicochemical properties, tea qualities, and tea nutrient contents. Graphs were prepared using SigmaPlot ver. 12.5 (Systat, Software, Inc., San Jose, CA).

RESULTS AND DISCUSSION

Basic Physicochemical Properties of Soil

Compared with CK, application of K (either SOP or POLY4) increased the content of soil available K but had no impact on N content in any year (Table 1). In addition, SOP and POLY4 application had no impact on P content in 2015, but significantly increased it in 2016 and 2017, a pattern consistent with that reported by Hu et al. (2012). There were two possible reasons for this: that balanced N, P, and K fertilization could increase microbial metabolic activity (Chu et al., 2007), which benefited active soil organic P compounds and increased soil available P; or that balanced fertilization reduced P adsorption capacity of soil and increased soil available P (Verma et al., 2005). The increase of soil available P ceased in SOP and POLY4 treatments in 2016, possibly because soil available P reached dynamic equilibrium in soil. Because POLY4 is richer in Ca, Mg, and S than SOP at the rate of 180 K_2O ha^{-1}, POLY4 increased the contents of exchangeable Ca and available S in soil in 2016 and 2017 compared with SOP and also that of exchangeable Mg in the 3 yr examined. Available S in soil was at high level and increased with time, which was attributed to serious acid rain in the study area (Liu et al., 2018). The K in POLY4 was readily available to tea plants as it is in SOP, as shown by the lack of difference in soil available K concentration in the experiment. This was also previously found in a column leaching study, in which the release rate of K from polyhalite powder was slightly higher than from soluble fertilizers (Barbarick, 1991).

Previous studies (Ruan et al., 2007; Yan et al., 2018) showed that tea is unique in that it requires acidic soil, and, in turn, makes soil more acidic—our study had similar results. Soil pH reduced in the three treatments during 2015 to 2017. First, soil acidification was partly attributed to organic acids and protons released from tea roots and deficiency in soil basic cations, such as Ca and Mg, in tea gardens (Yan et al., 2018). Second, the aluminum was absorbed and accumulated in tea leaves, and the biogeochemical cycling of aluminum in tea leaves can cause soil acidification (Song and Liu, 1990; Ding and Huang, 1991). In addition, the serious acid rain in south China is another possible reason for it (Liu et al., 2018). However, after 3 yr, soil pH decreased by 0.74 with SOP, by 0.49 with POLY4, and by 0.55 in CK. Thus, SOP application most lowered the soil pH, consistent with the results of Yan et al. (2016). It is possible that K application benefited tea growth and yield, and also enhanced aluminum uptake and accumulation in tea leaves, although aluminum in tea leaves and soil was not determined in our study. Similarly, compared with SOP, the significantly increased tea yield under POLY4 led to higher tea biomass, and the biogeochemical cycling of tea leaves have contributed to the soil pH decrease of 0.32 during 2016 to 2017, but a decrease of 0.08 occurred under SOP. After 3 yr, POLY4 application significantly slowed the acidification of soil compared with CK or SOP. A possible reason was that POLY4 application introduced large amounts of $CaSO_4$ and $MgSO_4$ to the soil and thus enhanced soil base cations (Yang et al., 2018). In addition, SO_4^{2-} has a high replacing power for OH^- groups on soil surfaces, and soil pH is usually raised slightly by SO_4^{2-} addition (Kiehl and Franco, 1984). Fung and Wong (2004) and Jayaganesh et al. (2006) respectively confirmed that $CaSO_4$ and $MgSO_4$ addition slightly increased soil pH, while Hailes et al. (1997) found that extractable Mg was positively correlated with soil pH. Although tea favors acidic soil (its optimal soil pH is 4.5–6.0), pH below 4.0 inhibits plant growth and increases solubility of some metals. Yaylali and Tüysüz (2009) found negative correlations between soil pH and availability of Pb, Zn, Mn, and Al elements in tea plants. The metals among these absorbed by tea plants may entail a risk to human health (Seenivasan et al., 2008; Li et al., 2016; de Oliveira et al., 2018). These aspects make POLY4 particularly suitable for tea plantations. However, given high soil available S content after 3 yr of POLY4 application, it might be better to apply POLY4 together with other K fertilizers.

Table 2. Effects of source of K on N, P, K, Ca, and Mg contents of tea leaves.

Year	Treatment†	N content		P content		K content		Ca content		Mg content		S content	
		Spring tea	Summer tea	Spring tea	Summer tea	Spring tea	Summer tea	Spring tea	Summer tea	Spring tea	Summer tea	Spring tea	Summer tea
		g kg^{-1}											
2016	Control	65.5 ± 1.04a‡	30.9 ± 0.40a	5.50 ± 0.24a	3.20 ± 0.07a	16.4 ± 0.18a	17.3 ± 0.47a	3.26 ± 0.03a	0.49 ± 0.04a	1.61 ± 0.07ab	3.41 ± 0.05a	nd§	nd
	SOP	65.6 ± 0.87a	32.9 ± 1.35a	5.61 ± 0.13a	3.62 ± 0.13b	17.3 ± 0.30b	20.1 ± 0.38b	3.36 ± 0.12a	0.48 ± 0.04a	1.57 ± 0.09a	3.48 ± 0.05a	nd	nd
	POLY4	66.5 ± 0.44a	34.1 ± 2.41a	5.80 ± 0.02a	3.73 ± 0.13b	17.2 ± 0.49b	20.4 ± 1.16b	3.99 ± 0.12b	0.52 ± 0.03a	1.82 ± 0.06b	3.81 ± 0.10b	nd	nd
2017	Control	56.6 ± 2.01a	39.2 ± 0.51a	5.58 ± 0.05a	3.43 ± 0.04a	14.7 ± 0.25a	17.6 ± 0.71a	3.23 ± 0.09a	1.20 ± 0.01a	1.40 ± 0.04a	2.11 ± 0.07a	1.92 ± 0.04a	1.99 ± 0.05a
	SOP	58.2 ± 2.41a	39.2 ± 0.64a	5.63 ± 0.07a	3.60 ± 0.08b	15.8 ± 0.17b	19.5 ± 0.17b	3.35 ± 0.10a	1.31 ± 0.06a	1.27 ± 0.05a	2.20 ± 0.14a	2.10 ± 0.04b	2.25 ± 0.06b
	POLY4	60.3 ± 1.99a	40.5 ± 0.99a	5.81 ± 0.22a	3.69 ± 0.03b	16.2 ± 0.43b	20.2 ± 0.53b	4.07 ± 0.04b	1.64 ± 0.01b	1.89 ± 0.08b	2.85 ± 0.07b	2.15 ± 0.05b	2.27 ± 0.02b

† SOP, commercial sulfate of potash; POLY4, polyhalite.

‡ Values are mean ± standard deviation ($n = 3$). Means with the same letters in the same column in same year are not significantly different by Tukey test ($P > 0.05$).

§ nd, not determined.

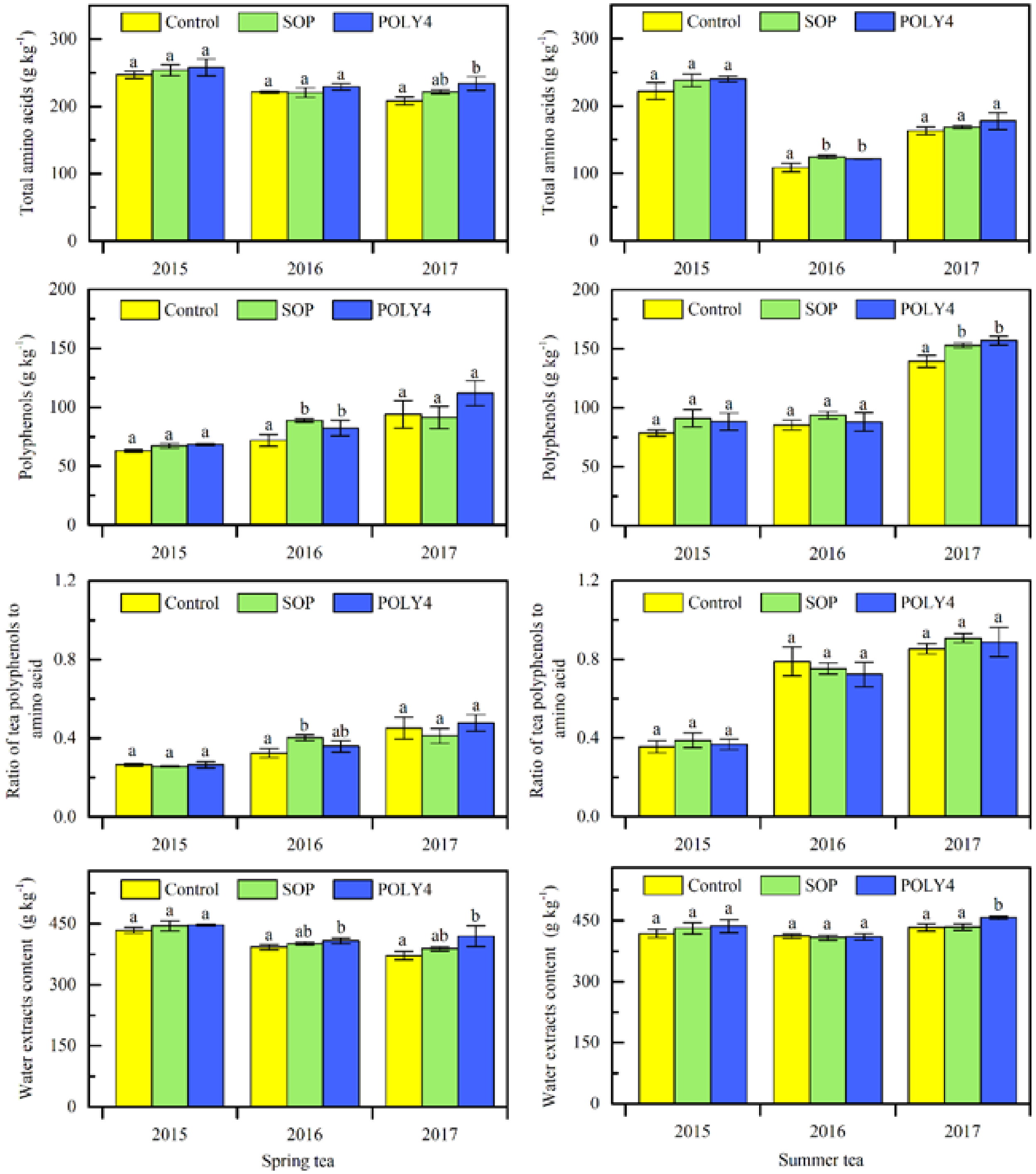

Fig. 1. Quality of tea as affected by the source of K (SOP, commercial sulfate of potash; POLY4, polyhalite). Values are mean ± standard deviation ($n = 3$). Means with the same letters in the same subpicture of the same year are not significantly different by Tukey test ($P > 0.05$).

Contents of N, P, K, Ca, Mg, and S in Tea Leaves

Table 2 shows that N contents of spring and summer tea were similar for all the three treatments in 2016 and 2017 ($P > 0.05$). Application of K led to significantly higher K content in the produce compared to that in the produce from CK, but the source of K made no difference. The N and P contents were higher in spring than in summer tea in the three treatments. There were three reasons for this: (i) Top dressing occurred before spring tea harvest, and more N and P from the fertilizer were absorbed in spring compared to summer tea. (ii) Buds and two adjacent expanding leaves are harvested for spring tea, compared with buds and one adjacent leaf for summer tea. (iii) The pruning practice between spring and summer tea each year in the study would change the N and P distribution in tea plants. There were similar phenomena for Ca and Mg contents in spring and summer tea. The much higher Mg content in summer than in spring tea was possibly due to a high migration rate of Mg (Gerendás and Führs, 2013). The K application had no effect on P content of the spring harvest but increased the P content of the summer harvest significantly, compared with CK ($P < 0.05$). This difference may be due to two reasons: first, the early growth was not lacking in access to P as the full dose of P had been applied as a basal dose in all the three treatments; and second, with the passage of time, the proportion of P in available form may have

Table 3. Pearson's correlation coefficients for spring tea quality and tea nutrient contents in 2017.

	Tea AA†	Tea WEC	Tea Po	Po/AA	Tea N	Tea P	Tea K	Tea Ca	Tea Mg	Tea S
Tea AA	1									
Tea WEC	0.883**	1								
Tea Po	0.613	0.833**	1							
Po/AA	0.209	0.542	0.900**	1						
Tea N	0.645	0.490	0.275	–0.006	1					
Tea P	0.799**	0.880**	0.684*	0.403	0.598	1				
Tea K	0.904**	0.753*	0.323	–0.100	0.526	0.617	1			
Tea Ca	0.810**	0.838**	0.748*	0.478	0.508	0.604	0.714*	1		
Tea Mg	0.697**	0.735*	0.697*	0.473	0.583	0.649	0.547	0.917**	1	
Tea S	0.536	0.426	–0.046	–0.346	0.434	0.277	0.775*	0.332	0.091	1

* Significant at $P < 0.05$; ** Significant at $P < 0.01$.

† AA, total amino acids; WEC, water extract content; Po, polyphenols; Po/AA, the ratio of tea polyphenols to amino acids.

decreased due to sorption, precipitation, and microbial immobilization (Zhu et al., 2018). In addition, imbalanced N, P, and K fertilization can hinder absorption of P by tea plants (Shen et al., 2007; Hu et al., 2012). Because of Ca, Mg, and S addition through POLY4, soil exchangeable Ca and Mg, and available S contents under POLY4 significantly increased in 2016, 2015, and 2016, respectively, compared with SOP. However, the contents of these nutrients in tea leaves were somewhat different to those in soil. The Ca and Mg addition from POLY4 resulted in higher contents of Ca and Mg in tea leaves, but there was no difference on S content in tea leaves for the two K fertilizers treatments during the 3 yr because of sufficient soil available S.

Quality of Tea

The flavor of tea is attributed to the balance between astringency and umami (savory) (Lin et al., 2012). Total free amino acids are the major contributors to the umami of green tea, and high concentrations of tea polyphenols lead to strong astringency; therefore, the ratio of tea polyphenols to amino acids is taken as an indicator of quality and is considered more important than absolute concentrations (Ruan et al., 1999; Hu et al., 2001). In addition, water extract content is also a parameter of tea quality, especially taste, as it contains amino acids and polyphenols (Yao et al., 2006; Xu et al., 2018).

Although our method of measuring total amino acids differed from some other studies (Lin et al., 2012; Ruan et al., 2012), there was a positive and significant relationship between the two amino acids (Supplemental Fig. S1). Thus, our higher content of total amino acids in tea leaves may have included a relatively higher proportion of total free amino acids. The spring harvest was generally of higher quality than the summer harvest as the amino acid contents were higher, the polyphenol contents were lower, and the ratio of polyphenols to amino acids was also lower (Fig. 1). Such spring tea accordingly fetches a higher price in China and in other countries, an observation partly in accord with that of Huang et al. (2007). The tea quality parameters varied among years. In 2015, the source of K had no effect on total amino acids, polyphenols, the ratio of polyphenols to amino acids, or water extract content compared with CK. However, in 2016, compared with CK, application of K fertilizers significantly increased amino acids in the summer harvest and polyphenols in the spring harvest, but had no impact on amino acids in spring and on polyphenols and water extract content in summer, because of low rainfall in 2016. The ratio of polyphenols to amino acids in the spring harvest in the SOP treatment and the water extract content in spring in the POLY4 treatment was higher than the corresponding values in CK. In 2017, application of K increased content of polyphenols in the summer harvest but had no impact on polyphenols in spring, amino acids in summer, and the ratio of polyphenols to amino acids in both spring and summer harvests. There was no significant difference between sources of K on quality parameters, except in 2017 when water extract content in the summer harvest was greater for the POLY4 treatment. Thus, POLY4 had no adverse effect on quality in the 3 yr.

Some research has proposed that tea is a calcifuge and acidophilic plant (Karak et al., 2017). To determine the effects Ca addition from POLY4 on tea quality, correlation analysis was conducted between spring tea parameters and soil parameters (Tables 3 and 4). Tea total amino acids was positively and significantly correlated with tea water extract content in spring and summer tea, and the two parameters both were significantly and positively correlated with tea P, tea K, tea Ca, and tea Mg in spring tea. Similar correlations existed in summer tea. Tea polyphenols in spring tea were significantly and positively correlated with tea P, Ca, and Mg, but significantly and positively correlated with tea P, K, Ca, Mg, and S in summer tea. It was concluded that tea qualities were generally positively and significantly correlated with tea P, K, Ca, and Mg. The K fertilization increased soil P availability (Table 1) and resulted in tea P being positively associated with tea quality. Tea K was positively correlated with tea Ca, and tea Ca was also significantly and positively correlated with tea Mg in spring and summer tea. This means that Ca supplied from POLY4 did not restrict K uptake by tea plants in the experiment, although Fung and Wong (2004) and Willson (1975a) found that uptake of K by tea can be inhibited by excess Ca in soil. The Pearson's correlation analysis confirmed that Ca addition from POLY4 did not harm tea quality, but was positively correlated with it. Willson (1975a, 1975b) demonstrated an antagonistic effect between K and Mg in soil, but we found no such phenomenon in our experiment. Tea quality generally showed no significant correlations with tea S, and S was not a factor affecting tea quality in this soil of high available S content.

Yield and Economic Benefits

Tea yield and the economic benefit from using POLY4 will primarily determine whether POLY4 is likely to be adopted. The three treatments showed no significant difference in yield, either seasonal or in total, in 2015 ($P > 0.05$; Fig. 2), probably because the soil was rich in K following many years of K

Table 4. Pearson's correlation coefficients for the summer and total tea yield, soil properties, summer tea quality, and summer tea nutrient content in 2017.

	TY†	SY	Tea AA	Tea WEC	Tea Po	Po/AA	Tea N	Tea P	Tea K	Tea Ca	Tea Mg	Tea S	pH	Soil N_A	Soil P_A	Soil K_A	Soil Ca^{2+}_E	Soil Mg^{2+}_E	Soil S_A
TY	–																		
SY	0.947**	–																	
Tea AA	0.661	0.692*	–																
Tea WEC	0.745*	0.559	0.694*	–															
Tea Po	0.934**	0.897**	0.507	0.639	–														
Po/AA	0.364	0.294	–0.410	0.018	0.577	–													
Tea N	0.606	0.477	0.353	0.649	0.404	0.090	–												
Tea P	0.909**	0.854**	0.595	0.594	0.784*	0.275	0.473	–											
Tea K	0.907**	0.932**	0.816**	0.709*	0.886**	0.164	0.434	0.775*	–										
Tea Ca	0.923**	0.804**	0.666*	0.823**	0.783*	0.199	0.714*	0.852**	0.780*	–									
Tea Mg	0.821**	0.728*	0.605	0.773*	0.679*	0.138	0.798**	0.648	0.685*	0.933**	–								
Tea S	0.890**	0.943**	0.579	0.480	0.917**	0.425	0.344	0.820**	0.920**	0.712*	0.575	–							
pH	0.519	0.347	0.099	0.502	0.428	0.359	0.690*	0.480	0.297	0.720*	0.717*	0.367	–						
Soil N_A	0.268	0.521	0.372	–0.056	0.371	0.031	–0.061	0.123	0.555	–0.015	0.019	0.568	–0.324	–					
Soil P_A	0.631	0.711*	0.395	0.262	0.678*	0.341	0.125	0.472	0.588	0.397	0.393	0.579	–0.170	0.432	–				
Soil K_A	0.816**	0.876**	0.486	0.438	0.803**	0.393	0.457	0.764*	0.832**	0.577	0.458	0.902**	0.218	0.633	0.575	–			
Soil Ca^{2+}_E	0.805**	0.698*	0.525	0.670*	0.659	0.204	0.688*	0.702*	0.596	0.912**	0.910**	0.557	0.686*	–0.146	0.476	0.394	–		
Soil Mg^{2+}_E	0.816**	0.641	0.561	0.840**	0.631	0.137	0.806**	0.769*	0.619	0.963**	0.918**	0.528	0.789*	–0.217	0.225	0.441	0.883**	–	
Soil S_A	0.961**	0.880**	0.722*	0.850**	0.832**	0.198	0.756*	0.859**	0.878**	0.930**	0.858**	0.787*	0.532	0.216	0.525	0.780*	0.796*	0.875**	–

* Significant at $P < 0.05$; ** Significant at $P < 0.01$.

† TY, total tea yield in 2017; SY, summer tea yield in 2017; N_A, available N; P_A, available P; K_A, available K; Ca^{2+}_E, exchangeable Ca; Mg^{2+}_E, exchangeable Mg; S_A, available S; AA, total amino acid; WEC, water extract content; Po, polyphenols; Po/AA, the ratio of tea polyphenols to amino acids.

application (Ruan et al., 2013) and also because tea, a perennial evergreen shrub, stores large amounts of K (Yan et al., 2018). Once the initial reserves of K were depleted, both POLY4 and SOP significantly increased yield in the summer of 2016 by 20.6 and 26.4%, respectively, and thus correspondingly for that whole year by 16.1 and 15.4% compared with CK. In 2016, total tea yield between SOP and POLY4 did not significantly differ. Compared with CK, POLY4 and SOP increased the yield in spring by 43.0 and 21.4%, in the summer of 2017 by 58.4 and 46.4%, and for the whole year by 46.9% and 27.6%, respectively. In 2017, yield of spring tea and the whole year under POLY4 application was significantly higher by 17.8 and 15.1% compared with SOP, respectively.

The Pearson's correlation coefficients for summer and total tea yield in 2017, tea nutrient contents, and soil properties are shown in Table 4. The total tea yield in 2017 was positively and significantly correlated with soil exchangeable Ca and Mg, available K and S contents, and tea P, K, Ca, and Mg contents, whereas summer tea yield was additionally correlated with soil available P. Because there was no difference in K input and soil available K content between SOP and POLY4 treatments, and the S content in soil was high, higher tea yields under POLY4 than under SOP in 2017 was attributed to enhanced soil fertility by Ca and Mg addition from POLY4 fertilizer. First, high soil basic cations (Ca, K, and Mg) from POLY4 fertilizer could increase soil fertility, which benefits tea growth (McKenzie et al., 2004). Second, Ca is an essential nutrient for tea growth. Although Karak et al. (2017) proposed that tea was a calcifuge plant, the Ca effects on tea plant largely depend on Ca form and application rate. Willson (1975a) demonstrated that only excess Ca application inhibited K uptake in tea. Fung and Wong (2004) found that among five forms of Ca, $CaSO_4$ application led to higher tea biomass than no Ca treatment, but CaO and $Ca(OH)_2$ application reduced biomass, because CaO and $Ca(OH)_2$ increased soil pH and reduced the aluminum content in tea, and aluminum was useful for root growth. It can be concluded that Ca was beneficial to tea growth, but the adverse effect of some Ca compounds on tea growth was mainly associated with the concomitantly and largely increased soil pH. In addition, Mg is involved in several physiological and biochemical processes, such as being the central element in chlorophyll, and acting as a cofactor in various enzymatic processes associated with photosynthesis, respiration, and energetic metabolism (Hermans and Verbruggen, 2005). Fertilization with Mg has been proposed to increase tea yield in many studies (Jayaganesh et al., 2006; Ruan et al., 1999, 2012). Because the same K application rate was applied in the two K fertilization treatments, high amount of Ca and Mg from POLY4 fertilizers contributed to higher tea yield in POLY4 treatment. Although MOP is a major K fertilizer in crop production, it was not included in this experiment mainly because some studies proposed detrimental effects of Cl from MOP on accumulation of free amino acids or theanine in young tea shoots (Ruan et al., 1998, 2007). In addition to potential Cl detrimental effects, POLY4

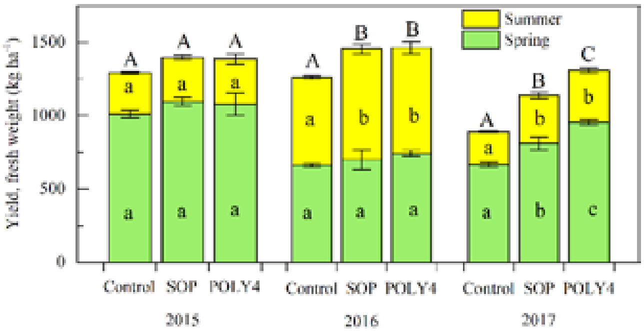

Fig. 2. Effects of K fertilizer application on tea yield (SOP, commercial sulfate of potash; POLY4, polyhalite). Values are mean ± standard deviation (n = 3). Means with the different upper-case letters for the same year are significantly different on total tea yield, and the different lower-case letters mean significantly different on yields of spring or summer tea in each year by Tukey test ($P < 0.05$).

may have performed better than MOP because POLY4 contains $CaSO_4$ and $MgSO_4$.

The economic benefits of using POLY4 and SOP fertilizers are compared in Table 5. Because there are many factors involved in tea net revenue—including cost of fertilizers (N, P, and K); labor costs of fertilization, irrigation, and tea harvesting; and income of different seasonal tea (yield and price)—we focused on the relative increased net revenue between POLY4, SOP, and CK. As all other costs such as pesticides, N and P fertilizers, and irrigation were equal for all treatments, the difference in revenue between POLY4, SOP, and CK was mainly because of costs of K fertilizer, labor for harvesting, and yield. The price of SOP and POLY4 was US\$0.48 kg^{-1} and \$0.20 kg^{-1}, respectively, according to the SOP distributor in China and one manager of Sirius Minerals Plc, respectively. The cost of labor for picking tea was \$5.12 kg^{-1} fresh tea, and the price of fresh spring and summer tea was \$18.08 kg^{-1} and \$12.05 kg^{-1} fresh tea, respectively. Both POLY4 and SOP resulted in higher revenue than CK in the 3 yr (Table 5), and the difference between K fertilization and CK increased with experiment duration—greater by \$1062 ha^{-1} and \$800 ha^{-1} in 2015, respectively, and correspondingly \$1382 ha^{-1} and \$1625 ha^{-1} in 2016, and \$2381 ha^{-1} and \$4363 ha^{-1} in 2017. Compared to SOP fertilization, POLY4 fertilization achieved higher revenue by \$243 ha^{-1} in 2016 and by \$1982 ha^{-1} in 2017 but achieved lower revenue in 2015 (–\$262 ha^{-1}). Thus, overall, POLY4 application resulted in both higher yields and higher economic benefit compared to the conventional source of K, namely SOP.

CONCLUSIONS

Fertilization with K is essential to tea yield and quality. Compared with CK, the SOP and POLY4 application significantly increased tea yield, economic benefits and soil fertility starting from the second year (2016) in the experiment. After 3 yr of application, there were higher soil exchangeable Ca and Mg and available S contents for POLY4 than SOP, and soil acidification was significantly reduced for POLY4 because of high addition of Ca and Mg, compared to SOP—this decrease is desirable for sustainable production of tea. Compared to SOP, POLY4 significantly increased both tea yield and economic benefits, especially in the third year (2017), because it added large quantities of $CaSO_4$ and $MgSO_4$ to soil. Although the two treatments did not differ significantly in tea quality, the correlation analysis demonstrated that tea quality was generally positively and significantly correlated with K, Ca, Mg, P, and S contents in tea leaves. Consequently, POLY4 was suitable as a source of K in tea plantations, and this is the first such demonstration. Given high S and Ca contents in POLY4, it might be a potential source to be used in future together with other K fertilizers.

ACKNOWLEDGMENTS

This work was financially supported by Sirius Minerals Plc, the National Key Research and Development Program of China (grant no. 2016YFD0200900), the Youth Foundation of Sichuan Academy of Agricultural Sciences (grant no. 2018QNJJ-017), the Fund for Excellent Papers of Sichuan Academy of Agricultural Sciences (grant no. 2018LWJJ-006), and the Open Foundation of State Key Laboratory of Soil and Sustainable Agriculture of China (grant no. Y20160021). We thank International Science Editing (http://www.internationalscienceediting.com) for editing this manuscript.

SUPPLEMENTAL MATERIAL

Supplemental material is available with the online version of this article. The supplement contains Supplemental Fig. S1, Relation

Table 5. Economic benefits of applying POLY4 and SOP.

Year	Treatment†	K fertilizer price	K fertilizer cost‡	Tea yield		Fresh tea income§	Labor cost of tea-picking¶	Tea income minus cost of K and labor	Increased revenue compared to CK#
				Spring tea	Summer tea				
		\$ kg^{-1}	\$ ha^{-1}	kg ha^{-1}		\$ ha^{-1}			
2015	Control	–	0	1,012	280	21,671	6,615	15,056	–
	SOP	0.48	172.80	1,095	303	23,449	7,158	16,118	1,062
	POLY4	0.20	257.14	1,078	309	23,214	7,101	15,856	800
2016	Control	–	0	662	597	19,163	6,446	12,717	–
	SOP	0.48	172.80	698	754	21,706	7,434	14,099	1,382
	POLY4	0.20	257.14	742	719	22,079	7,480	14,342	1,625
2017	Control	–	0	668	223	14,765	4,562	10,203	–
	SOP	0.48	172.80	810	326	18,573	5,816	12,584	2,381
	POLY4	0.20	257.14	955	353	21,520	6,697	14,566	4,363

† SOP, commercial sulfate of potash; POLY4, polyhalite.

‡ K fertilizer cost = K fertilizer price × K fertilizer product dose (CK was 0, SOP and POLY4 were both 180 kg K_2O ha^{-1} in both treatments).

§ Fresh tea income = (yield in spring × price in spring) + (yield in summer × price in summer).

¶ Labor cost of tea-picking = total tea yield in a year × local price of tea-picking labor (\$5.12 kg^{-1}).

Increased revenue compared to CK is the difference between SOP or between POLY4 and CK.

between total free amino acids and total amino acids according to different methods.

REFERENCES

Albadarin, A.B., T.D. Lewis, and G.M. Walker. 2017. Granulated polyhalite fertilizer caking propensity. Powder Technol. 308:193–199. doi:10.1016/j.powtec.2016.12.004

Alekseeva, T., A. Alekseev, R.K. Xu, A.Z. Zhao, and P. Kalinin. 2011. Effect of soil acidification induced by a tea plantation on chemical and mineralogical properties of Alfisols in eastern China. Environ. Geochem. Health 33:137–148. doi:10.1007/s10653-010-9327-5

Barbarick, K.A. 1991. Polyhalite application to sorghum-sudangrass and leaching in soil columns. Soil Sci. 151:159–166. doi:10.1097/00010694-199102000-00005

Chinese National Standard GB/T 5009.124. 2016. Determination of amino acid in food. (In Chinese.) China Food and Drug Administration, Beijing, China.

Chinese National Standard GB/T 8305. 2013. Tea-Determination in of water extracts content. (In Chinese.) Standardization Administration of the People's Republic of China, Beijing, China.

Chinese National Standard GB/T 8313. 2008. Determination of total polyphenols and catechins content in tea. (In Chinese.) Standardization Administration of the People's Republic of China, Beijing, China.

Chu, H.Y., X.G. Lin, T. Fuji, S. Morimoto, K. Yagi, J.L. Hu, and J.B. Zhang. 2007. Soil microbial biomass, dehydrogenase activity, bacterial community structure in response to long-term fertilizer management. Soil Biol. Biochem. 39:2971–2976. doi:10.1016/j.soilbio.2007.05.031

de Oliveira, L.M., S. Das, E.B. da Silva, P. Gao, J. Gress, Y.G. Liu, and L.Q. Ma. 2018. Metal concentrations in traditional and herbal teas and their potential risks to human health. Sci. Total Environ. 633:649–657. doi:10.1016/j.scitotenv.2018.03.215

Ding, R.X., and X.A. Huang. 1991. Biogeochemical cycle of aluminium and fluorine in a tea garden soil system and its relationship to soil acidification. (In Chinese). Turang Xuebao 28:229–236.

Fei, T.Y., J. Fei, F. Huang, T.P. Xie, J.F. Xu, Y. Zhou, and P. Yang. 2017. The anti-aging and anti-oxidation effects of tea water extract in *Caenorhabditis elegans*. Exp. Gerontol. 97:89–96. doi:10.1016/j.exger.2017.07.015

Fraps, G.S., and H. Schmidt. 1932. Availability to plants of potash in polyhalite. Texas Agricultural Experiment Station Bulletin, 449. Texas A&M University, College Station, TX.

Fung, K.F., and M.H. Wong. 2004. Application of different forms of calcium to tea soil to prevent aluminum and fluorine accumulation. J. Sci. Food Agric. 84:1469–1477. doi:10.1002/jsfa.1842

Gerendás, J., and H. Führs. 2013. The significance of magnesium for crop quality. Plant Soil 368:101–128. doi:10.1007/s11104-012-1555-2

Hu, C., J.Y. Song, J. Li, C.Q. Liu, X.E. Tao, Y.F. Chen, Y. Qiao, S.L. Li, and D.H. Liu. 2012. Profile distribution of soil available phosphorus and available potassium concentrations and crop grain yields in long-term fertilization experiment. (In Chinese). Ecol. Environ. Sci. 21:673–676.

Han, W., S.J. Kemmitt, and P.C. Brookes. 2007. Soil microbial biomass and activity in Chinese tea gardens of varying stand age and productivity. Soil Biol. Biochem. 39:1468–1478. doi:10.1016/j.soilbio.2006.12.029

Hailes, J.K., R.L. Aitken, and N.W. Menzies. 1997. Magnesium in tropical and subtropical soils from north-eastern Australia. I. Magnesium fractions and interrelationships with soil properties. Soil Res. 35:615–627. doi:10.1071/S96081

Hermans, C., and N. Verbruggen. 2005. Physiological characterization of Mg deficiency in Arabidopsis thaliana. J. Exp. Bot. 56:2153–2161. doi:10.1093/jxb/eri215

Hu, Q., J. Xu, and G. Pan. 2001. Effect of selenium spraying on green tea quality. J. Sci. Food Agric. 81:1387–1390. doi:10.1002/jsfa.952

Huang, Y., J. Sheng, F. Yang, and Q.H. Hu. 2007. Effect of enzyme inactivation by microwave and oven heating on preservation quality of green tea. J. Food Eng. 78:687–692. doi:10.1016/j.jfoodeng.2005.11.007

Jayaganesh, S., S. Venkatesan, and V.K. Senthurpan. 2006. Impact of different sources and doses of magnesium fertilizer on biochemical constituents and quality parameters of black tea. Asian J. Biochem. 6:273–281.

Karak, T., K. Bora, R.K. Paul, S. Das, P. Khare, A.K. Dutta, and R.K. Boruah. 2017. Paradigm shift of contamination risk of six heavy metals in tea (*Camellia sinensis*, L.) growing soil: A new approach influenced by inorganic and organic amendments. J. Hazard. Mater. 338:250–264. doi:10.1016/j.jhazmat.2017.05.036

Karak, T., R.K. Paul, R.K. Boruah, I. Sonar, B. Bordoloi, A.K. Dutta, and B. Borkotoky. 2015. Major soil chemical properties of the major tea-growing areas in India. Pedosphere 25:316–328. doi:10.1016/S1002-0160(15)60016-9

Kiehl, J.C., and O. Franco. 1984. Efeito do gesso industrial solve alguns componentes da fertilidade do solo. Solo 76:11–16.

Kemp, S.J., F.W. Smith, D. Wagner, I. Mounteney, C.P. Bell, C.J. Milne, C.J.B. Gowing, and T.L. Pottas. 2016. An improved approach to characterize potash-bearing evaporite deposits, evidenced in North Yorkshire, United Kingdom. Econ. Geol. 111:719–742. doi:10.2113/econgeo.111.3.719

Lepeshkov, I.N., and A.N. Shaposhnikova. 1958. Natural polyhalite salt as a new type of potassium-magnesium-boron fertilizer. Udobr. Uzozh. 11:33–35.

Li, S.Y., X. Wu, H. Xue, B.J. Gu, H. Cheng, J.M. Zeng, Y.G. Peng, and J. Chang. 2011a. Quantifying carbon storage for tea plantations in China. Agric. Ecosyst. Environ. 141:390–398. doi:10.1016/j.agee.2011.04.003

Li, P.S., Y.S. Wu, X. Du, H.L. Shan, H. Yang, and Q. Tang. 2011b. Productive characteristics of famous green tea of 5 introduced tea plant cultivars in West Sichuan province. (In Chinese.) J. Sichuan Agric. Univ. 3:334–341.

Li, S.Y., H.X. Li, C.L. Yang, Y.D. Wang, H. Xue, and Y.F. Niu. 2016. Rates of soil acidification in tea plantations and possible causes. Agric. Ecosyst. Environ. 233:60–66. doi:10.1016/j.agee.2016.08.036

Li, T., H.Y. Wang, J. Wang, Z.J. Zhou, and J.M. Zhou. 2015. Exploring the potential of phyllosilicate minerals as potassium fertilizers using sodium tetraphenylboron and intensive cropping with perennial ryegrass. Sci. Rep. 5:9249. doi:10.1038/srep09249

Liu, X., Z.Y. Fu, B. Zhang, L. Zhai, M.J. Meng, J. Lin, J.Y. Zhuang, G.G. Wang, and J.C. Zhang. 2018. Effects of sulfuric, nitric, and mixed acid rain on Chinese fir sapling growth in Southern China. Ecotoxicol. Environ. Saf. 160:154–161. doi:10.1016/j.ecoenv.2018.04.071

Lin, Z.H., Y.P. Qi, R.B. Chen, F.Z. Zhang, and L.S. Chen. 2012. Effects of phosphorus supply on the quality of green tea. Food Chem. 130:908–914. doi:10.1016/j.foodchem.2011.08.008

Lu, L.K. 1999. Analytical methods of soil and agricultural chemistry. (In Chinese.) Chinese Agricultural Science and Technology Press, Beijing.

Lu, X.K., Q.G. Mao, F.S. Gilliam, Y.Q. Luo, and J.M. Mo. 2014. Nitrogen deposition contributes to soil acidification in tropical ecosystems. Glob. Change Biol. 20:3790–3801. doi:10.1111/gcb.12665

McKenzie, N., D. Jacquier, R. Isbell, and K. Brown. 2004. Australian Soils and Landscapes: An Illustrated Compendium. CSIRO, Collingwood, p. 62–64. doi:10.1071/9780643100732

Ogorodova, L.P., M.F. Vigasina, L.V. Mel'chakova, I.A. Bryzgalov, and D.A. Ksenofontov. 2016. Enthalpy of formation of natural polyhalite. Geochem. Int. 54:645–649. doi:10.1134/S0016702916070077

Panitkin, V.A. 1967. Effect of polyhalite on sandy loam soil. Agrokhimiya 1:81–84.

Rajan, J., and S.V. Anandhan. 2016. Influence of nitrogen and potassium on root nutrient and root CEC of different tea cultivars (*Camellia sinensis*, *C. assamica* and *C. assamica* spp. *Lasiocalyx*). Rhizosphere 1:36–44. doi:10.1016/j.rhisph.2016.07.004

Ruan, J.Y., J. Gerendás, R. Härdter, and B. Sattelmacher. 2007. Effect of nitrogen form and root-zone pH on growth and nitrogen uptake of tea (*Camellia sinensis*) plants. Ann. Bot. 99:301–310. doi:10.1093/aob/mcl258

Ruan, J.Y., L.F. Ma, and Y.Z. Shi. 2013. Potassium management in tea plantations: Its uptake by field plants, status in soils, and efficacy on yields and quality of teas in China. J. Plant Nutr. Soil Sci. 176:450–459. doi:10.1002/jpln.201200175

Ruan, J.Y., L.F. Ma, and Y. Yang. 2012. Magnesium nutrition on accumulation and transport of amino acids in tea plants. J. Sci. Food Agric. 92:1375–1383. doi:10.1002/jsfa.4709

Ruan, J.Y., X. Wu, and R. Härdter. 1999. Effects of potassium and magnesium nutrition on the quality components of different types of tea. J. Sci. Food Agric. 79:47–52. doi:10.1002/(SICI)1097-0010(199901)79:1<47::AID-JSFA172>3.0.CO;2-A

Ruan, J.Y., X. Wu, Y. Ye, and R. Härdter. 1998. Effect of potassium, magnesium and sulphur applied in different forms of fertilizers on free amino acid content in leaves of tea (*Camellia sinensis* L.). J. Sci. Food Agric. 76:389–396. doi:10.1002/(SICI)1097-0010(199803)76:3<389::AID-JSFA963>3.0.CO;2-X

Seenivasan, S., N. Manikandan, N.N. Muraleedharan, and R. Selvasunndaram. 2008. Heavy metal content of black teas from south India. Food Control 19:746–749. doi:10.1016/j.foodcont.2007.07.012

Shekhar, S., D. Mishra, A. Agrawal, and K.K. Sahu. 2017. Physical and chemical characterization and recovery of potash fertilizer from glauconitic clay for agricultural application. Appl. Clay Sci. 143:50–56. doi:10.1016/j.clay.2017.03.016

Shen, M.X., L.Z. Yang, Y.M. Yao, D.D. Wu, J.G. Wang, R.L. Guo, and S.X. Yin. 2007. Long-term effects of fertilizer managements on crop yields and organic carbon storage of a typical rice–wheat agroecosystem of China. Biol. Fertil. Soils 44:187–200. doi:10.1007/s00374-007-0194-x

Singh, A.K., and S.K. Pathak. 2018. Potassium in tea (*Camellia sinensis* (L) O. Kuntze) cultivation from soil to cup quality- A review. Agric. Rev. (Karnal) 39:40–47.

Song, M., and Y. Liu. 1990. Effect of biogeochemical cycle in tea garden on the soil acidification. (In Chinese.) Chaye Kexue 10:19–26.

Su, S.L., C. Wan, J. Li, X.F. Jin, J.H. Pi, Q.W. Zhang, and M. Weng. 2017. Economic benefit and ecological cost of enlarging tea cultivation in subtropical China: Characterizing the trade-off for policy implications. Land Use Policy 66:183–195. doi:10.1016/j.landusepol.2017.04.044

Venkatesan, S., S. Murugesan, V.K.S. Pandian, and M.N.K. Ganapathy. 2005. Impact of sources and doses of potassium on biochemical and green leaf parameters of tea. Food Chem. 90:535–539. doi:10.1016/j.foodchem.2004.05.014

Verma, S., S.K. Subehia, and S.P. Sharma. 2005. Phosphorus fractions in an acid soil continuously fertilized with mineral and organic fertilizers. Biol. Fertil. Soils 41:295–300. doi:10.1007/s00374-004-0810-y

Willson, K.C. 1975a. Studies on the mineral nutrition of tea V. Calcium. Plant Soil 43:295–307. doi:10.1007/BF01928493

Willson, K.C. 1975b. Studies on the mineral nutrition of tea VI. Magnesium. Plant Soil 43:309–316. doi:10.1007/BF01928494

Xu, Y., Y.T. Li, T. Bao, X.D. Zheng, W. Chen, and J.X. Wang. 2017. A recyclable protein resource derived from cauliflower by-products: Potential biological activities of protein hydrolysates. Food Chem. 221:114–122. doi:10.1016/j.foodchem.2016.10.053

Xu, Y.Q., W.B. Ji, P.G. Yu, J.X. Chen, F. Wang, and J.F. Yin. 2018. Effect of extraction methods on the chemical components and taste quality of green tea extract. Food Chem. 248:146–154. doi:10.1016/j.foodchem.2017.12.060

Yan, P., C. Shen, L.C. Fan, X. Li, L.P. Zhang, L. Zhang, and W.Y. Han. 2018. Tea planting affects soil acidification and nitrogen and phosphorus distribution in soil. Agric. Ecosyst. Environ. 254:20–25. doi:10.1016/j.agee.2017.11.015

Yan, Z., Z.Q. Gao, Z.H. Luo, X. Hu, L. Peng, and Q.R. Zeng. 2016. Effect of potassium fertilizers on the uptake of soil cadmium by flue-cured tobaccos. (In Chinese.) Environ. Chem. 35:1913–1920.

Yao, L.H., X. Liu, Y.M. Jiang, N. Caffin, B. D'Arcy, R. Singanusong, N. Datta, and Y. Xu. 2006. Compositional analysis of teas from Australian supermarkets. Food Chem. 94:115–122. doi:10.1016/j.foodchem.2004.11.009

Yang, X.D., K. Ni, Y.Z. Shi, X.Y. Yi, Q.F. Zhang, L. Fang, L.F. Ma, and J.Y. Ruan. 2018. Effects of long-term nitrogen application on soil acidification and solution chemistry of a tea plantation in China. Agric. Ecosyst. Environ. 252:74–82. doi:10.1016/j.agee.2017.10.004

Yaylali, G., and N. Tüysüz. 2009. Heavy metal contamination of soils and tea plants in the eastern Black Sea region, NE Turkey. Environ. Earth Sci. 59:131–144. doi:10.1007/s12665-009-0011-y

Zeng, L.T., Y. Zhou, X.M. Fu, X. Mei, S.H. Cheng, J.D. Gui, F. Dong, J.C. Tang, S.Z. Ma, and Z.Y. Yang. 2017. Does oolong tea (*Camellia sinensis*) made from a combination of leaf and stem smell more aromatic than leaf-only tea? Contribution of the stem to oolong tea aroma. Food Chem. 237:488–498. doi:10.1016/j.foodchem.2017.05.137

Zhao, H., J. Huang, Y. Li, X.W. Song, J.L. Luo, Z. Yu, and D.J. Ni. 2016. Natural variation of selenium concentration in diverse tea plant (*Camellia sinensis*) accessions at seedling stage. Sci. Hortic. (Amsterdam) 198:163–169. doi:10.1016/j.scienta.2015.11.026

Zhao, X.Y., L.Y. An, N. Liu, H.A. Yin, and M.L. Tang. 2011. Laboratory simulation of in-situ leaching of polyhalite. Proc. Earth Planet. Sci. 2:50–57. doi:10.1016/j.proeps.2011.09.009

Zhu, J., M. Li, and M. Whelan. 2018. Phosphorus activators contribute to legacy phosphorus availability in agricultural soils: A review. Sci. Total Environ. 612:522–537. doi:10.1016/j.scitotenv.2017.08.095