



Bark Effects on Stemflow Chemistry in a Japanese Temperate Forest I. The Role of Bark Surface Morphology

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Stemflow can be an important pathway for the drainage of precipitation and related solutes through tree canopies to forest soils. As stemflow must drain along bark surfaces, the effects of bark structure on stemflow chemical composition is merited. This study examines the relationship between stemflow chemistry and bark surface structure for six species of varying bark morphology (four deciduous broadleaf trees and two evergreen coniferous trees) at a montane and an urban site in Japan. Stemflow from smooth-barked species contained greater concentrations of solutes that appear to be rinsed from the stem surface (i.e., sea salt aerosols); while, rougher-barked tree species contained greater or less concentrations of solutes that appear to be leached (e.g., Ca²⁺) or taken-up (e.g., inorganic N) by the bark, respectively. Site-specific atmospheric environments also influenced the bark-stemflow chemistry relationships—where the greater elemental deposition in the urban plot generally resulted in greater stemflow chemistry than observed in the lower-deposition montane plot. Our results therefore suggest that the dynamics of dry deposition wash-off by stemflow, and the exchange of dissolved solutes between stemflow and the bark surface, are influenced by the surface structure of the bark and the site's atmospheric environment. Therefore, the interactions between bark surface structure and its surrounding atmospheric environment are important factors in the stemflow-related elemental cycling between the tree and precipitation.

Keywords: bark morphology, direct rainfall, inorganic carbon, mineral cycling, stemflow, throughfall

INTRODUCTION

As rainfall drains through tree canopies, it becomes an important nutrient input to the forest floor. This is because a portion of the rainwater contacts the leaves, branches, and epiphytes of the tree canopy, becoming throughfall (droplets that pass through, and drip from the canopy), and stemflow (a portion that travels over the bark surface to the base of the tree stem). Throughfall and stemflow become chemically altered as solutes are leached from the tree canopy surfaces, dry-deposited components are washed away by the draining rainwaters, and these

surfaces (and/or their resident flora and fauna) uptake or transform solutes within the rainwater (Ponette-González et al., 2020). In forests, compared to other vegetated landscapes, throughfall accounts for the majority of the rainfall (Lin et al., 2020), while stemflow from trees rarely exceeds 2% of total rainfall (Van Stan and Gordon, 2018). Stemflow's small proportion of the canopy rainwater balance, however, does not prevent it from contributing significantly to the net rainfall nutrient flux (Levia and Germer, 2015). Because stemflow can collect water from a large area of the tree canopy and spatially concentrate this water to a small area at the base of the tree, 10^{-2} – 10^1 m² tree⁻¹ (Van Stan and Allen, 2020), it can be a concentrated nutrient flux worthy of investigation in ecosystem ecology and hydrology disciplines (Levia and Germer, 2015; Van Stan and Gordon, 2018). In fact, several studies in various forest systems have found stemflow to be able to contribute 10–51% of various dissolved inorganic N fluxes per hectare, despite being <10% of gross rainfall (Liu et al., 2003; Pilegaard et al., 2003; Oziegbe et al., 2011; Germer et al., 2012). Thus, it is necessary to incorporate stemflow in investigations of forest water and nutrient cycling processes.

Stemflow can be highly enriched in a variety of solutes, including inorganic cations (e.g., K⁺, Na⁺, Ca²⁺, Mg²⁺, NH₄⁺), anions (e.g., Cl⁻, SO₄²⁻, NO₃⁻) (Parker, 1983), and dissolved organic C (DOC: Stubbins et al., 2020). The chemical composition of stemflow can differ substantially among tree species and across forest settings (e.g., Limpert and Siegert, 2019; Liu et al., 2019; Su et al., 2019). Variability in stemflow chemistry across forest settings may be related to differences in surrounding landscape properties (Rodrigo et al., 2003), and storm synoptic patterns (Siegert et al., 2017). Stemflow chemical variability across species at the same site, may be due to interspecific differences in canopy structures that alter the canopies' ability to capture and accumulate aerosols or change the solute leaching and uptake properties of canopy surfaces (Levia et al., 2011). As stemflow drains primarily down bark-covered canopy elements (like branches and stems), some past research examining interspecific differences in stemflow chemistry has focused on differences in bark external structure (Levia and Herwitz, 2005; McGee et al., 2019).

The structure of the bark surface can affect stemflow chemistry by altering the amount and type of dry deposited materials capture on bark (Xu et al., 2019), the corticolous epiphytic vegetation and microbes (Magyar, 2008; McDonald et al., 2017), the bark surface area for solute exchange (Whittaker and Woodwell, 1967), and the water storage and residence time in the canopy (Levia and Herwitz, 2005). Few studies explicitly examine the interaction between bark surface structure and stemflow chemistry. Levia and Herwitz (2005) conducted a comparative study of the chemical composition and water retention of stemflow in three broad-leaved trees with different bark morphologies (*Betula lenta* L., *Carya glabra* Mill., and *Quercus rubra* L.) with a purpose similar to that of the present study, and reported that capacity of bark water storage and stemflow chemistry were different among bark surface morphology. Limpert and Siegert (2019) examined stemflow for a wider suite of solutes, including macronutrient ions and

dissolved organic matter, and reported that thicker-barked (fire-tolerant) oaks produced more enriched stemflow than thinner-bark species, theoretically due to greater residence time. Su et al. (2019) reported the flux of inorganic ions in forests by stemflow analysis for deciduous trees with deeply furrowed bark and evergreen trees with smoother bark, finding that the stemflow from the deciduous trees was more chemically enriched due to greater surface area, contact time, and more dead materials in the rougher bark.

These studies show that the dynamics of bark surface-mediated solute exchange with stemflow can play a part in the nutrient cycle of forests. Thus, this study aims to build on these previous findings by conducting comparative evaluations of the impact of tree bark surface structure on stemflow chemistry (pH, dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, Mg²⁺, Ca²⁺, and K⁺) across multiple tree species situated within study sites that represent pristine (the Minami Alps mountains) and suburban (Tokyo) sites in Japan. Seven tree species representing a range of bark surface structure, from smooth-to-rough, were selected for the study (**Figure 1**). We expect to reject the null hypothesis: stemflow chemistry will not differ among species of differing bark surface structure. Results are expected to show that, for stemflow from smoother-to-rougher bark: (i) solutes predominantly originating from wash-off (like Na⁺, Cl⁻, and DOC) and leaching (like Mg²⁺, Ca²⁺, and K⁺) will become more concentrated; but (ii) solutes susceptible to being taken up or transformed by the bark surface (like NO₃⁻ and NH₄⁺) will become less concentrated. The relationships between stemflow solute concentrations and bark surface structure are also expected to (iii) differ between sites due to the influence of human activities and differences in rainfall itself.

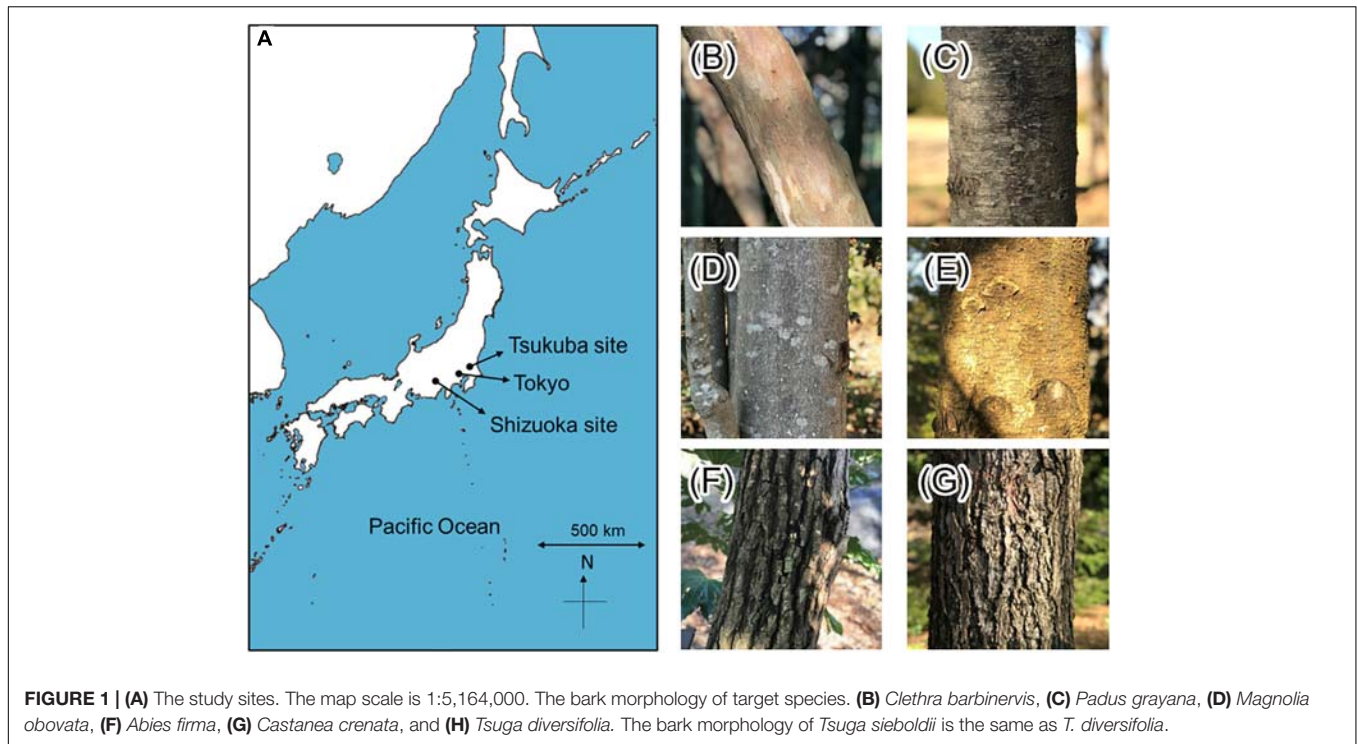
MATERIALS AND METHODS

Study Sites

The study sites included a montane, temperate forest in Tashiro of Shizuoka City, central Japan (the Shizuoka site, **Figure 1**); and, a suburban, temperate forest at the Tsukuba Experimental Forest, Mountain Research Center, University of Tsukuba, Japan (the Tsukuba site, **Figure 1**). For the Shizuoka site (35.307672 N, 138.199925 E), the elevation is 966 m a.s.l., the annual rainfall is 3110.1 mm, and the annual mean temperature is 11.4°C; the Tsukuba site (36.115326 N, 140.101504 E) has an elevation of 25 m a.s.l. and an annual rainfall of 1282.9 mm and an annual average temperature of 13.8°C (1981–2010 statistical period per AMeDAS: Japan Meteorological Agency, 2019). Canopy height of the study site at the Shizuoka site was approx. 20 m (Seino and Endoh, 2019), and at the Tsukuba site was almost the same as at the Shizuoka site (Seino, personal observations). The Shizuoka site is approx. 48 km far from the nearest coast, and the Tsukuba site is approx. 45 km away.

Study Species and Their Bark Structure

The tree species selected for this study included deciduous broadleaved and evergreen conifer species (**Figure 1**). The



selected deciduous broadleaved trees were found at both the montane and the suburban forest sites, and included *Clethra barbinervis* Siebold. & Zucc. (Japanese Clethra), *Padus grayana* Maxim. (Japanese bird cherry), *Magnolia obovata* Thunb. (Japanese whitebark magnolia), and *Castanea crenata* Siebold. & Zucc. (Japanese chestnut). The selected evergreen conifers included *Abies firma* Siebold. & Zucc. (momi fir) at both sites, *Tsuga sieboldii* Carr. (Japanese hemlock) only at Shizuoka site, and *Tsuga diversifolia* (Maxim.) Mast. (kometsuga) only at Tsukuba site. The bark types were categorized and photos showing the bark surface structure are provided in **Figure 1**. Both of the *Tsuga* species have a coarse, thick, exfoliating bark with fissures, where *T. diversifolia* has a vertically peeling bark structure while *T. sieboldii* has a square-like cracked and flaky bark structure. The bark of *A. firma* is scaly and has resinous blisters, which is smoother than the *Tsuga* species but rougher and thicker than many of the broadleaved species. *Clethra barbinervis* also has exfoliating bark, but it is the thinnest bark of all the study species—although the exfoliating bark flakes add a “mottling” to the surface. The bark of *P. grayana* is also quite smooth and thin; however, it is laterally marked by shallow lenticels. *Magnolia obovata* presents a stippled, gray bark that is moderately rough and thick, yet *C. crenata* has the thickest bark of all the study species and is deeply furrowed.

Rainfall and Stemflow Sampling

The study period ran from May to December 2018 with tree selection and equipment installation from May to August 2018. Sound individual trees with low inclination of the stem and little influence by other canopy cover or vine plants were selected as target trees. Rain and stemflow water samples were collected

and chemically analyzed for four storms. Sampling dates for the Shizuoka site were September 11, September 26, October 17, and November 29 2018; and, for the Tsukuba site were September 12, September 23, October 10, and December 9 2018 (**Figure 2**). We recognize that sampling only four storms limits the power of our analyses due to under-replication. These storms represent a range of their rainfall amount and duration.

Rainfall was collected by drilling a hole in the lid of a 5 L polyethylene tank and attaching a 9 cm diameter funnel. To prevent litter and insects from entering the funnel, a ping-pong ball was placed in the funnel, and the entire funnel was covered with a net (**Figure 3**). One gauge each was installed at the Shizuoka and Tsukuba site. Stemflow was collected by installing collars to capture waters draining down the outside of the stem which drained these waters to polyethylene collection bins using the following methods. First, to prevent rainwater from seeping out of the collar, the lichen on the stems where the collar was to be installed was removed, and paraffin wax was applied. Second, a 5 cm wide, 5 mm thick rubber roll was placed beneath and fixed to the tree stem to divert the water draining down the bark surface into a 25 cm wide, 3 mm thick rubber sheet with a water intake valve at the bottom (which was wrapped around the stem). Instant adhesive and silicone for caulking were used to bond the rubber to the tree stem. One end of a vinyl hose, with water pipe sealing tape wrapped around both ends, was inserted into the outer rubber collar where the water outlet was located, and the other end was connected to a 5 L polyethylene tank with a hole drilled in the lid. Finally, rubber bands were used to secure the poly tank to the stem (**Figure 3**). In total, 148 water samples were obtained during the study period for the six target species with three replicate trees per species (Shizuoka

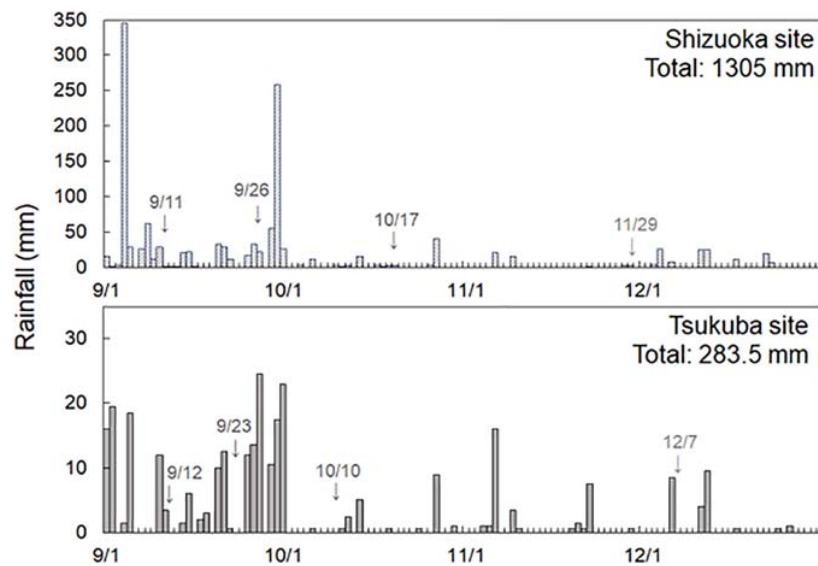


FIGURE 2 | Rainfall and sampling date from 1 September to 31 December 2018 at the Shizuoka and Tsukuba sites. Rainfall data at the Shizuoka site is from the Water Information System of the Ministry of Land, Infrastructure, Transport and Tourism (<http://www1.river.go.jp/cgi-bin/SrchRainData.exe?ID=105031285523010&KIND=1&PAGE=0>, last viewed 3 December 2019), and at Tsukuba site is measured by the meteorological station of the Tsukuba Experimental Forest, Mountain Science Center, University of Tsukuba.

site: rainfall ($n = 4$), stemflow ($n = 72$); Tsukuba site: rainfall ($n = 4$), stemflow ($n = 68$). Between storms, rainfall and stemflow samplers were rinsed by DW after every sampling. Gauges were sampled within 24 h of the end of a storm event.

Chemical Analysis

Each water sample was filtered through a $0.45 \mu\text{m}$ filter (GLCTD-MCE2545, Shimadzu GLC, Tokyo), then the following chemical parameters were measured: pH, dissolved organic carbon (DOC), inorganic carbon (DIC), and inorganic ion concentrations (Cl^- , NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , Mg^{2+} , Ca^{2+} , and K^+). A total organic carbon analyzer (TOC-V CSH, Shimadzu, Kyoto) was used to measure DOC and DIC. Inorganic ion concentrations were measured using ion chromatography (Prominence series, Shimadzu, Kyoto). Calibration curves were prepared using five mixed standard solution for the peak area of each ion, and quantification was performed after confirming their correlation coefficients.

Statistical Analysis

Statistical analyses were performed using R version 3.4.2 (R Core Team, 2017) with a multiple-way analysis of variance (ANOVA) for comparing effects between sites and tree species, and differences among species were compared by one-way ANOVA and Tukey's multiple comparisons for chemistries of rainfall and stemflow. Heteroscedasticity of each values was checked by Bartlett's test before ANOVA. To evaluate patterns within the several stemflow chemical parameters, altogether, we applied a Principal Components Analysis on a matrix of log-transformed solute concentration data using the online ClustVis

application¹ by Metsalu and Vilo (2015). ClustVis was chosen, not only due to its simplicity, but due to its ease of reproducibility.

RESULTS

Rainfall Solute Concentrations

Rainwater solute concentrations were all lower for the montane site (Shizuoka) compared to the urban site (Tsukuba) (**Figure 4**). Mean ($\pm\text{SD}$) of rainwater concentrations in DOC, Cl^- , SO_4^{2-} , Na^+ , Mg^{2+} , Ca^{2+} , and K^+ for Shizuoka were $1.4 \pm 0.6 \text{ mg L}^{-1}$, $6.3 \pm 4.1 \mu\text{mol L}^{-1}$, $2.2 \pm 1.6 \mu\text{mol L}^{-1}$, $7.7 \pm 2.9 \mu\text{mol L}^{-1}$, $1.3 \pm 0.9 \mu\text{mol L}^{-1}$, $0.1 \pm 0.2 \mu\text{mol L}^{-1}$, and $1.3 \pm 2.2 \mu\text{mol L}^{-1}$, respectively. For the Tsukuba site, rainwater concentrations of the same solutes were larger and more variable, being $4.4 \pm 3.8 \text{ mg L}^{-1}$, $46.7 \pm 31.9 \mu\text{mol L}^{-1}$, $9.0 \pm 3.4 \mu\text{mol L}^{-1}$, $40.5 \pm 27.6 \mu\text{mol L}^{-1}$, $6.7 \pm 3.0 \mu\text{mol L}^{-1}$, $9.2 \pm 4.0 \mu\text{mol L}^{-1}$, and $7.4 \pm 4.9 \mu\text{mol L}^{-1}$, respectively. For the N solutes, all observations were below observation limits in rain samples from the montane Shizuoka site; while mean ($\pm\text{SD}$) NO_3^- and NH_4^+ concentrations for the urban site were $13.4 \pm 9.1 \mu\text{mol L}^{-1}$ and $16.8 \pm 15.3 \mu\text{mol L}^{-1}$, respectively. Thus, all chemical parameters measured in rainfall, barring pH, were statistically significant between sites (unpaired t -test, $p > 0.05$) for pH, $p < 0.05$ for excepted pH).

Stemflow Chemical Variability Across Species of Differing Bark Roughness

Across the study species, there was a general trend of decreasing stemflow pH with increasing bark roughness, from 6.0 ± 0.4

¹<https://biit.cs.ut.ee/clustvis/>

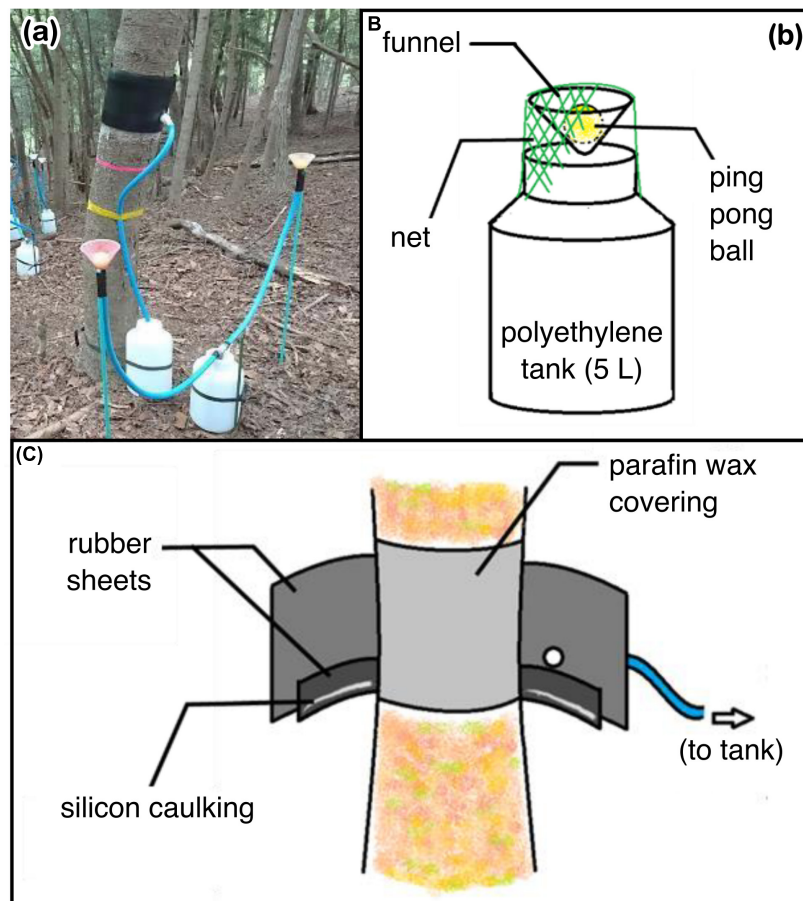


FIGURE 3 | Photograph and diagrams of the water sampling system. **(a)** Photograph of the implementation on *Abies firma* at the Shizuoka site, **(b)** diagram of water collector for direct rainfall, **(c)** water collection system for stemflow.

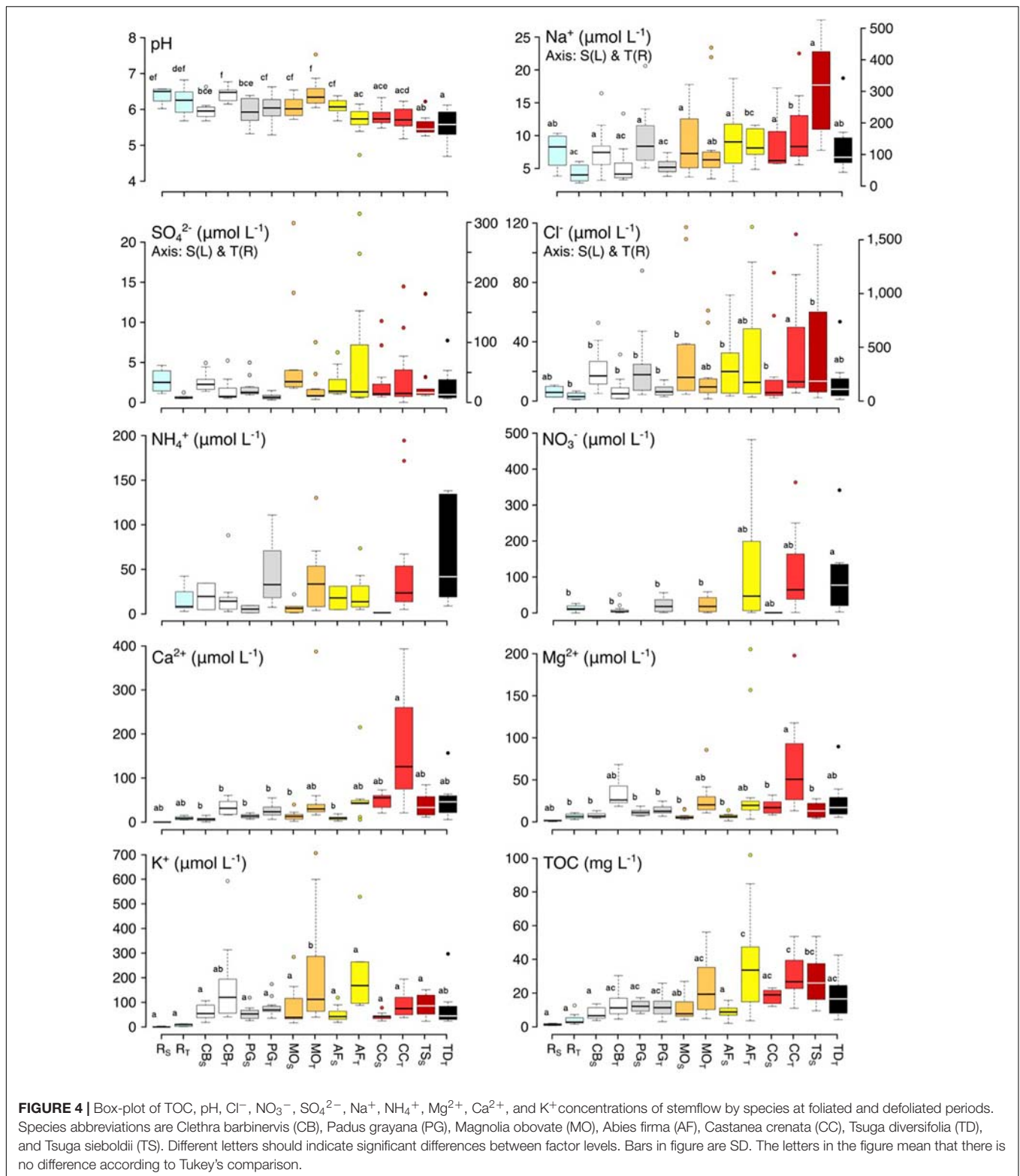
for smooth-barked (broadleaved) species like *C. barbinervis* and *P. grayana* to 5.5 ± 0.4 for rough-barked (evergreen) species like *T. sieboldii* and *T. diversifolia* (**Figure 4**). Na^+ , SO_4^{2-} , Cl^- , and Ca^{2+} also showed the same trend as pH, and species and site effects were detected for Na^+ and Ca^{2+} [Two-way ANOVA, $F_{(5, 132)} = 3.07$, $p < 0.001$ for Na^+ ; $F_{(0.129)} = 2.81$, $p < 0.001$ for Ca^{2+}], while only site effects were detected for SO_4^{2-} [Two-way ANOVA, $F_{(1, 132)} = 3.07$, $p < 0.001$]. Deep teared bark type as *C. crenata* especially high in Ca^{2+} . Patterns of NH_4^+ were low in *C. barbinervis* and coniferous species. In broad-leaved trees, the concentration in *C. crenata* stemflow tended to be higher at both the Shizuoka and Tsukuba sites, while for conifers, the stemflow of *T. sieboldii* was higher than that of *A. firma* at the Shizuoka site, whereas that of *T. diversifolia* was lower than that of *A. firma* at the Tsukuba site (**Figure 4**).

Principal Components Analysis: Stemflow Chemical Variability Across Species and Sites

Further exploration of the several stemflow chemical parameters across species and sites was done by applying a principal

components analysis (**Figure 5**). Two principal components were selected that represented 76% of the variability in the stemflow chemistry dataset [64% in component 1 (PC1) and 12% in component 2 (PC2)]. Individual stemflow observations per species at each site (faded-color symbols) were widely distributed across the principal components space (**Figure 5**). The mean principal component scores for each species' stemflow chemistry at each site (represented by solid-colored symbols) show more distinctly how these observations amalgamated into two broad clusters, based primarily on site-specific differences (rather than species- or bark-related differences). The loadings plot (lower right of **Figure 5**) indicates that primarily “washed off” solutes (i.e., Na^+ , Cl^- , SO_4^{2-}) may explain the site-related clustering. This major site-related clustering based on solutes that were likely washed-off agrees with the rainfall data showing that the urban (Tsukuba) site had significantly higher rainwater solute concentrations compared to the montane (Shizuoka) site (**Figure 4**).

Secondarily, the loadings plot indicates that interspecies variability within these site-specific PCA clusters spread out in relation to their primarily “exchanged” solute concentrations (i.e., DOC, Ca^{2+} , K^+ , and Mg^{2+}). The ordering of species



with differing bark structure along the PCA plot's representation of solute "exchange" differs between sites. For the montane site, where solute deposition contributions were lower, the roughest-barked tree species, both broadleaved and needleleaved

(*C. crenata*, *T. sieboldii*) plotted high on PC2 (**Figure 5**). A cluster of smoother-barked tree species at Shizuoka, also both broadleaved and needleleaved (*C. barbinervis*, *A. firma*, *M. obovata*) is observed in the negative domain of PC2. The

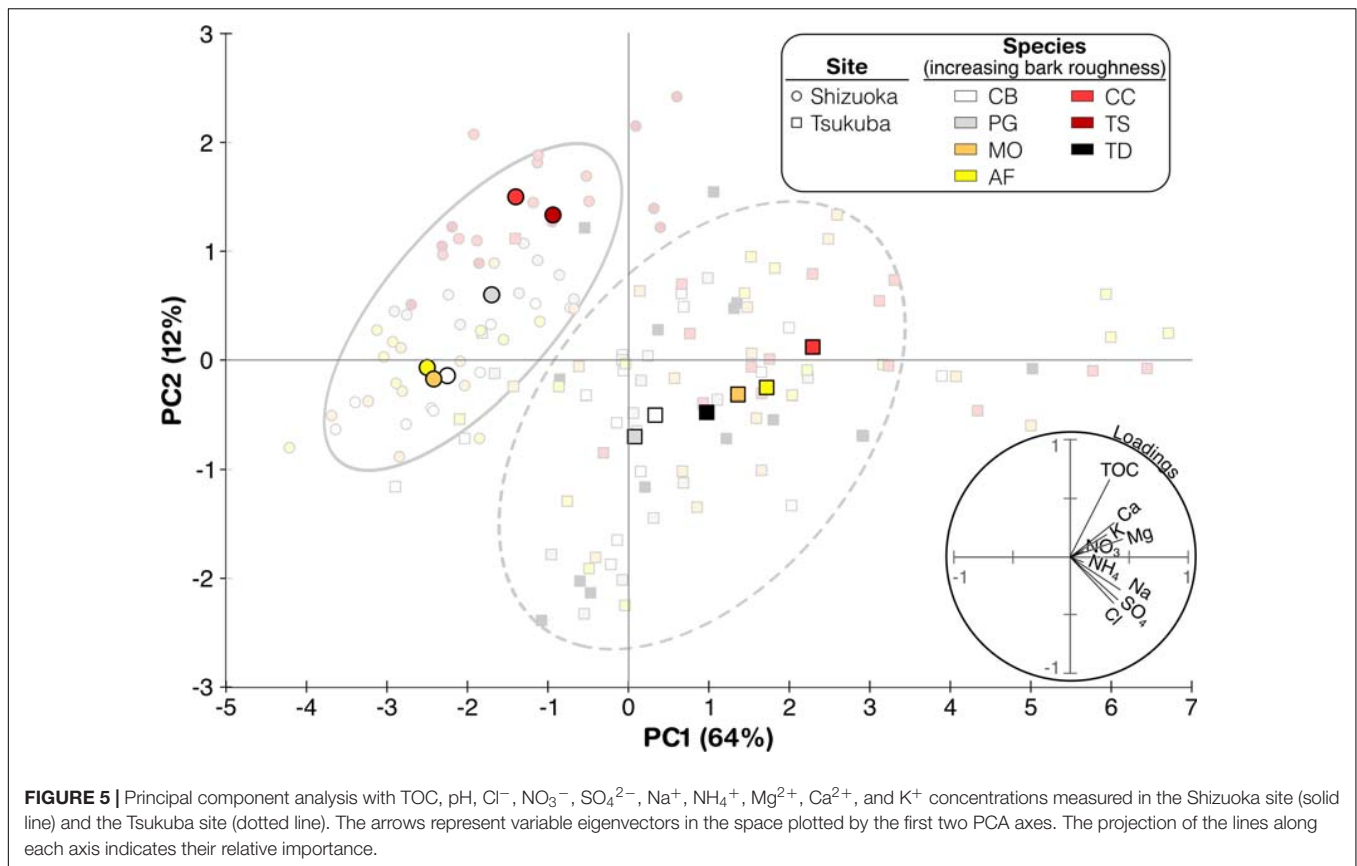


FIGURE 5 | Principal component analysis with TOC, pH, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, Mg²⁺, Ca²⁺, and K⁺ concentrations measured in the Shizuoka site (solid line) and the Tsukuba site (dotted line). The arrows represent variable eigenvectors in the space plotted by the first two PCA axes. The projection of the lines along each axis indicates their relative importance.

smooth-barked tree with deep lenticels, *P. grayana*, however, plotted intermediately between these other Shizuoka clusters on PC2. Interestingly, at the high-deposition urban site, Tsukuba, the interspecies variability within the PCA cluster is more evenly spaced along a continuum of smooth-to-rough bark (Figure 5). Although *T. diversifolia* is a rougher bark, its peeling bark structure is qualitatively smoother than *T. sieboldii* (see Figure 1), which may explain its location within the PCA space, nearby to the other more moderately rough barked species (*A. firma*, *M. obovata*). Thus, this PCA plot suggests that deposition chemistry is the principal driver of stemflow chemistry in this study; moreover, the site with higher deposition chemistry (Tsukuba) also produced a more evenly spaced solute “exchange” continuum along bark roughness (Figure 5).

DISCUSSION

Stemflow Chemical Variability Across Bark Roughness

Stemflow solute concentration and composition varied significantly across the bark roughness continuum represented by our seven study tree species, including both broadleaved and evergreen species (Figure 1). Thus, these results reject the null hypothesis that stemflow chemistry will not differ among species of differing bark surface structure. Relationships along the smoother-to-rougher bark continuum for stemflow chemistry

were consistent across sites, where solutes predominantly originating from atmospheric deposition and wash-off (like Na⁺, Cl⁻, and DOC) and leaching (like Mg²⁺, Ca²⁺, and K⁺) generally increased with increasing bark roughness (Figure 4). Indeed, the PCA plot for stemflow data from the site with higher deposition, the urban Tsukuba site, showed this continuum relatively clearly (Figure 5). Reported solute concentrations fall within the range of those reported in previous stemflow literature (Parker, 1983; Levia and Germer, 2015; Van Stan and Stubbins, 2018). The higher stemflow solute concentrations from rough-barked trees, like *C. crenata* and *T. sieboldii*, also agree with the highly enriched stemflow solutions observed from similarly rough barked tree species, like *Quercus* species (Levia and Herwitz, 2005; Van Stan et al., 2017), *Cyclobalanopsis* species (Su et al., 2019), *Fraxinus pennsylvanica* (Schooling et al., 2017), *Liriodendron tulipifera* (Levia et al., 2011). Therefore, in combination with previous results, our study supports the conceptual model that bark structure may decrease bulk stemflow solute concentration by influencing both (a) the amount of materials captured by bark and (b) the amount of stemflow available to dilute those washed off (and exchanged) materials (Van Stan and Gordon, 2018).

On the other hand, stemflow solutes susceptible to being taken up or transformed by the bark surface (like NO₃⁻ and NH₄⁺) displayed more complex responses across the species with changing bark roughness. In some cases, concentrations increased for smoother-barked trees, like mean NH₄⁺ was higher

for *P. grayana* than (the very rough-barked) *C. crenata* at the Tsukuba site (Figure 4). We note that concentrations of NO_3^- and NH_4^+ were low or almost undetectable for stemflow from many species and many storms (Figure 4). And that the loadings plot in the PCA shows a very small effect of N ion concentrations on overall patterns of stemflow chemistry (Figure 5). The generally low concentrations of these solutes may be due to strong uptake, as several studies have reported net uptake of both NH_4^+ and NO_3^- across forest sites, for throughfall as well as stemflow (Parker, 1983; Kloeti et al., 1989; Crockford et al., 1996; Cavalier et al., 1997; André et al., 2008). The instances of high inorganic N concentrations (higher than rainfall N concentrations) for some species at the urban Tsukuba site—especially those with moderate-to-rough bark—may reflect the amount of N build-up within these barks due to the urban deposition of particulates (i.e., Xu et al., 2019) rather than increased leaching. Although, we acknowledge the possibility of canopy N losses due to urban impacts on its nutrient intrasystem cycling (i.e., Decina et al., 2018). There was no difference in pH between the bark types. In the case of broad-leaved species, the pH of the stemflow of *C. crenata* was low, while that of *B. grossa*, which has the same coarse longitudinally split bark, was higher in Takenaka et al. (1998) (*C. crenata*, 5.61; *Quercus crispula* Blume, 5.79 at Aichi, central Japan) and Tsukahara (1994) (*C. crenata*, 5.6; *Q. crispula*, 6.2; at Yamagata, northern Japan), suggesting species specificity due to another factors (regional differences and so on) than the bark characters.

Comparison of Stemflow Chemistry Across Sites

For stemflow, the Shizuoka (montane) and Tsukuba (urban) sites showed very different trends in solute concentrations and solute composition. NO_3^- concentrations at the Shizuoka site were not sufficiently high to be considered, while at the Tsukuba site, concentrations were high in most samples (Figure 4). This difference may be due to the urban atmospheric deposition at Tsukuba compared to Shizuoka (Lovett et al., 2000). The stemflow of *C. crenata*, *A. firma*, and *T. sieboldii* tended to be relatively high, and the stemflow of *A. firma* in particular increased rapidly. The concentration of Na^+ was 10–20 times higher at the Tsukuba site than at the Shizuoka site. In contrast to the Shizuoka site, there was no difference between species at the Tsukuba site. NH_4^+ was hardly detected in samples at the Shizuoka site but was detected in many samples at the Tsukuba site. Although the concentrations of Mg^{2+} and Ca^{2+} were remarkably higher at the Tsukuba site than at the Shizuoka site, the trends at each site were similar (Figure 4). There was a common tendency for lower concentrations of direct rainfall and higher concentrations of *C. crenata* in stemflow relative to that of *C. barbinervis*, *P. grayana*, and *M. obovata*. In the stemflow of conifers at the Shizuoka site, *A. firma* was higher, while *A. firma* was higher at the Tsukuba site. For K^+ , the results were similar to those of Mg^{2+} and Ca^{2+} , while bark type had no effect, except for stemflow in *M. obovata*, and stemflow in *C. barbinervis*. Katagiri (1977) reported high Mg^{2+} and Ca^{2+} content in the trunk of *C. crenata* and high K^+ content in the leaves of *C. barbinervis*,

suggesting that the nutrient content in each layer of the tree is involved in the dynamics of leaching substances.

Overall, the major differences between the Shizuoka and Tsukuba sites are thought to be differences in component concentrations due to differences in rainfall (Figure 4), and the presence or absence of nitrogen components due to differences in human activities such as traffic volume. The concentrations of Mg^{2+} , Ca^{2+} , and K^+ for TOC and inorganic ions in Oka et al. (2019) suggested that organic acids are leaching along with these base cations, while the current results suggest that the counterparts to organic acids are mainly Mg^{2+} and Ca^{2+} , which are influenced by bark. Besides, the concentrations of nitrogen and K^+ obtained from the Tsukuba site suggest that NH_4^+ and K^+ moving in conjunction. One reason for this is that the Tsukuba site is close to a farm, so components from the application of fertilizer were dispersed and detected in the stem stream. Besides, a comparison of the leaf-dressing and leaf-declining seasons showed a tendency for higher concentrations in the leaf-declining season, which may be due to the greater precipitation in summer than in winter (Eaton et al., 1973; Su et al., 2019).

An interesting cross-site finding, from the PCA plot, was that the solutes generally considered as exchangeable (DOC, K^+ , etc.), showed little trend across the species of varying bark roughness at the montane site. However, there was a clear “lining up” of the species’ stemflow chemistry (with regard to highly exchangeable solutes) along a general bark roughness continuum for the urban site, where deposition was much higher (Figure 5). We believe that this finding is interesting, because one would expect that higher deposition of soluble elements to these tree canopies would “mask” any potential trends in bark solute exchange—yet, the alignment along these loadings for PC2 is apparent (Figure 5). Is this an indication that high-deposition environments impact, not only the capture and washoff of solutes, but the exchange of related solutes? This is a reasonable question as urban atmospheric deposition is known to impact canopy physiology (Calfapietra et al., 2015). Alternatively, the trend observed in the PCA related to bark roughness and exchangeable solutes may have little-to-do with increased bark-water-solute exchange, and more-to-do with the increased capture of soluble aerosols by the more effective (rougher) “bark traps” for particulates (Magyar, 2008; Xu et al., 2019).

Comparison Between Conifers and Broad-Leaved Trees

Many studies have found a tendency of low pH in coniferous stemflows (Sassa et al., 1990; Levia and Germer, 2015). The results also showed a tendency to lower stemflow, especially in two *Tsuga* species. The pH of *C. crenata* and *A. firma* also tends to decrease in response to stemflow. It has been reported that stemflow in broad-leaved trees inhibits the acidification of soil during the process of rainfall reaching the soil (Sassa and Hasegawa, 1992; Neary and Gizyn, 1994). This may explain why this effect is lower in *C. crenata* and conifer leaves, which are less able to repel rainfall and stay in the tree canopy due to their hard and well-developed cuticle, and why it tends to be

different in coarse-barked *Q. crispula* and *J. mandshurica*, as mentioned in the comparison with previous studies (Shibata and Sakuma, 1996; Levia and Herwitz, 2005; Oka et al., 2019). In terms of TOC, SO_4^{2-} , Mg^{2+} , and Ca^{2+} , the relationship between *M. obovata* and *C. crenata* at the Shizuoka site corresponded to the relationship between *A. firma* and two *Tsuga* species, suggesting that there is also a bark influence between conifers and broad-leaved trees. On the other hand, in stemflow at the Shizuoka site, Cl^- was higher in *M. obovata* than in *C. crenata*, and that in *A. firma* was lower than in *T. sieboldii*; however, at the Tsukuba site, Cl^- was lower in *M. obovata* than in *C. crenata*, and that in *A. firma* was higher than that in *T. diversifolia*, which was the complete opposite of the trend for conifers and broad-leaved trees. This is because the amount of stock of dry deposition on trees differs due to the difference in rainfall between sites, and the amount of stock is low in *M. obovata* and easy to retain in *C. crenata*. Rainfall capture by canopy leaves were different broad-leaved trees and conifers, and this was affected to stemflow chemistry (Shibata and Sakuma, 1996; Abbasian et al., 2015; Su et al., 2019). Besides, the leaf area of *Magnolia* and *Castanea* is relatively large, even as compared to other broad-leaved trees (White, 1983), and conifers have small needle leaves.

CONCLUSION

The results of this study agree with research at other sites that bark surface structure is related to stemflow chemistry. Additionally, this relationship between bark surface structure and stemflow chemistry at our sites depended on the tree canopies external environment (specifically, whether the atmospheric deposition environment was montane or urban). Site-specific differences appear to be driven by increased wet deposition (more concentrated rainfall chemistry) at the urban site compared to the montane site. The retention capacity of (assumed) dry deposited elements was different between study trees with smooth and rough barks—and their mobilization by stemflow depended on the rainfall amount at each site. At the Shizuoka site, where there is more (chemically dilute) rainfall, the smooth bark type, with its thin bark structure without grooves, appears to immediately rinse the bark of soluble minerals. On the other

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hand, the coarse bark type, with multi-layered and wavy surface structures, permitted the bark to store substantial amounts of soluble materials that became available during the stemflow process in the Shizuoka site. Our results support a growing body of literature that suggest bark surface structure can affect the dynamics of elemental cycling in the forest ecosystem by influencing stemflow chemistry.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author/s.

AUTHOR CONTRIBUTIONS

AO, YE, and TS collected samples. AO and JT carried out chemical analyses. All authors contributed to the writing of the manuscript, conceived and designed the research plan.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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