1	Rainfall redistribution in subtropical Chinese forests changes over 22
2	years
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#### 15 Abstract

Rainfall redistribution through the vegetation canopy plays a key role in the 16 hydrological cycle. Although there have been studies on the heterogeneous patterns of 17 rainfall redistribution in some ecosystems, the understanding of this process in different 18 stages of forest succession remains insufficient. Therefore, this study investigated the 19 change tendency of rainfall redistribution and rainwater chemistry in a subtropical 20 succession forest area in South China, based on 22 years (2001-2022) of monitoring 21 740 valid rainfall events. Results showed that at the event scale throughfall ratio showed 22 23 in order broadleaf forest (BF) < mixed forest (MF) < pine forest (PF), and stemflow ratio showed in order BF > MF > PF. At the interannual scale, throughfall and stemflow 24 of forests experienced a decreasing followed by an increasing from 2001–2022 (except 25 stemflow of the pine forest), similar with the trend of open rainfall (except the mutation 26 time). The variability of throughfall presented in order MF (CV, 9.7%)  $\leq$  PF (11.8%)  $\leq$ 27 BF (16.4%), and variability of stemflow presented in order MF (CV, 38.6%)  $\leq$  PF (50.9) 28 29 < BF (56.2%). This suggested that changes of throughfall and stemflow in the broadleaf forest were greater than those in the mixed forest and pine forest over time. Besides, 30 the difference of rainwater chemistry fluxes (TN, TP and K<sup>+</sup>) among the three forest 31 32 types were found and they changed in different order over time. On average, TN and TP fluxes of throughfall presented in order BF < MF < PF, while K<sup>+</sup> flux of throughfall 33 presented in order BF > MF > PF. Stemflow chemical fluxes varied less among forest 34 types and/or over time, though tree species exactly contribute to differences in stemflow 35 chemistry. The above results indicated that the patterns of rainfall redistribution 36 changed over time and showed characteristic variation driven by rainfall and forest 37 factors. This study provided insight into the rainfall redistribution process by linking 38 the long-term changing of rainfall pattern and subtropical forest succession sequence. 39 40 Keyword: throughfall, stemflow, variability, forest types, long-term

#### 41 1. Introduction

In recent years, there has been on-going concern about the potential impacts of 42 climate change on forest ecosystems, particularly in terms of rainfall input associated 43 to water resource (Reynaert et al., 2020; Grossiord et al., 2017; Bruijnzeel et al., 2011; 44 Leuzinger and Körner, 2010). Numerous studies have documented rainfall regimes and 45 their effect on the water cycle in different regions of the world, including spatial and 46 temporal changes in the amount, intensity, and frequency (Brasil et al., 2018; Ponette-47 González et al., 2010). Meanwhile, these variables in rainfall refer to the redistribution 48 49 of rainfall into canopy interception, throughfall and stemflow, being important components of terrestrial ecosystems hydrological processes (Germer et al., 2010; 50 Levia and Frost, 2006; Loustau et al., 1992). Rainfall redistribution patterns can impact 51 the biogeochemistry cycle by affecting soil moisture distribution, which in turn affects 52 the activity of soil microorganisms that decompose organic matter (Tonello et al., 2021a; 53 Junior et al., 2017; Van Stan II and Pypker, 2015). The study of Sun et al. (2023) verified 54 that throughfall reduction significantly affected soil carbon cycle in a subtropical forest. 55 Therefore, understanding the roles of rainfall redistribution in the water cycle is 56 essential. 57

58 Rainfall redistribution, as the partitioning into interception loss, throughfall and stemflow, is an important hydrological process that regulates water and nutrient cycling 59 60 in forest ecosystems. Interception loss refers to the part of the event rainfall intercepted by the canopy, accounting for about 10%-30% of gross rainfall depending on the 61 studied forest canopy, such as shrub (Zhang et al., 2015), mixed broadleaf (Yan et al., 62 2003), pine (Loustau et al., 1992). Later on, the remaining rainwater reaches the ground 63 either as throughfall or stemflow. Throughfall is a critical component of rainfall 64 redistribution, and it on average contributes to approximately 60%-90% of the gross 65 rainfall on the floor in forests, shrubland or cropland. (Zhang et al., 2023; Zhang et al., 66 2021; Brauman et al., 2010; Marin et al., 2000). Raindrops coalesce or splash on canopy 67 leaf surfaces, generating spatially different throughfall volume and raindrop kinetic 68 energies which can be larger or lower than that of open rainfall (Levia et al., 2019; 69 70 Goebes et al., 2015). Stemflow, the left rainwater flowing bottomwards along the plant stem or trunk, often accounts for only a small proportion (0-12%) of rainfall (Niu et al., 71 72 2023; Yue et al, 2021; Llorens and Domingo 2007). Nevertheless, stemflow inputs can be important as hot spots for near-trunk soils, inducing water and nutrient enrichment 73 and deep infiltration, but also erosion (Zhao et al., 2023; Llorens et al., 2022). It can 74

funnel more water than open rainfall on an equivalent area and contributes to 10% of the annual soil water input (Levia and Germer, 2015; Chang and Matzner, 2000). Throughfall and stemflow restrict water input to the soil layer, thereby affecting soil moisture conditions, runoff generation and water and nutrient cycling (Lian et al., 2022; Lacombe et al., 2018; Klos et al., 2014).

The proportions of rainfall redistribution are generally driven by meteorological 80 conditions (e.g., rainfall amount, intensity, duration) and vegetation cover (e.g., canopy 81 structures, tree characteristics) (Tonello et al., 2021a; Sun et al., 2018; Mużyło et al., 82 83 2012; Nanko et al., 2006). For meteorological conditions, numerous studies have documented that throughfall volume and stemflow volume increase with increasing 84 gross rainfall and intensity (Ji et al., 2023; André et al., 2011). The ratios of throughfall 85 and stemflow were both characterized with logarithmically increasing with rainfall, 86 tending to be quasi-constant for heavy rainfall events (Zhang et al., 2021; Liu et al., 87 88 2019). This was also synchronously related to the gradual saturation of the canopy which limited the ratios of rainwater partitioning (Carlyle-Moses et al., 2004). Besides, 89 90 differences of water volume spatially exist from place to place. The spatial variability (expressed as a coefficient of variation) of throughfall volume is generally higher for 91 small rainfall events (< 10 mm) than that for heavy rainfall events (Germer et al., 2006; 92 Price et al., 1997). 93

94 Rainfall redistribution among different plant species can vary significantly due to differences in the structure and characteristics of their canopies. In special, some of the 95 key factors determine the redistribution of rainfall, for example, leaf area index (LAI), 96 97 leaf shapes and orientations can affect the amount of intercepted loss and throughfall 98 (Zhang et al., 2021; Goebes et al., 2015; Keim et al., 2006). The diameter at breast 99 height (DBH), bark type and orientation of trunks/stems and branches influence the 100 amount of stemflow (Levia et al., 2015; Livesley et al., 2014; Germer et al., 2010). For each of the rainfall partitioning fluxes, their responses to the influential predictors often 101 show high variation. A modelling study of rainfall partitioning in China explained that 102 throughfall was best represented by mean tilt angle (MTA), followed by DBH. 103 104 Subsequently, DBH was the dominant predictor for stemflow, followed by LAI and bark texture (Zhang et al., 2023). Due to these factors, rainfall redistribution presented 105 different degrees of spatial variability. This variability (expressed as coefficient of 106 variation) decreased with increasing rainfall amount and intensity, consequently 107 tending to be quasi-constant (Germer et al., 2006). Besides, at interannual scale, ratios 108

of rainfall redistribution are driven by annual canopy structures. The study of Niu et al.
(2023) documented that annual throughfall ratio gradually increased, while annual
stemflow ratio and interception loss ratio decreased with increasing thinning intensity
in shrub plantation. Meanwhile, annual changes of rainfall events (amount and intensity)
reinforced the time instability of throughfall spatial variability (Rodrigues et al., 2022).
Overall, the rainfall-canopy interactions play a key role in rainfall redistribution
processes and further affect the water cycle in many ecosystems.

Vegetation canopy is the functional interface between ecosystem and atmospheric 116 117 wet deposition (Van Stan II and Pypker, 2015). The leaf and trunk/stem, acting as a filter, alter rainwater chemical concentrations via leaching and depositing processes. As 118 a result, throughfall and stemflow exhibit high chemical concentrations compared to 119 the open rainfall (Jiang et al., 2021; Zimmermann et al., 2007). For instance, in a Chinese 120 pine plantation, the volume weighted mean concentrations of NH4<sup>+</sup> and NO3<sup>-</sup> in 121 throughfall were significantly higher than those in open rainfall (Wang et al., 2023). 122 123 Stemflow ion fluxes (e.g., K<sup>+</sup>) from deciduous tree species were greater than those for evergreen tree species because of the differences in bark morphology and branch 124 125 architecture (Su et al., 2019). Moreover, it is also common that throughfall and stemflow chemistry fluctuated seasonality with the shifts in rainfall regime and leaf 126 127 growth (Turpault et al., 2021; Siegert and Levia, 2014; Staelens et al., 2007). A large number of elements required by plants are mainly N, P, K, Ca and Mg. In general, N, K 128 129 and Ca are the most important inputs to forest ecosystem, and P is the least. Phosphorus (P) is considered to be a limiting nutrient element in tropical and subtropical forests. 130 The long-term productivity of vegetation depends on the input of atmospheric P. 131 Besides, the increasing trend in seasonal drought and atmospheric nitrogen (N) 132 deposition in subtropical areas of China were reported (Zhou et al., 2011), and may 133 134 inhibit the growth of plant and affect productivity and functioning of forest ecosystems (Wu et al., 2023; Borghetti et al., 2017). Therefore, the important significance of 135 atmospheric precipitation to the ecosystem can also be seen from the amount of element 136 cycling, and canopy leaching also plays an important role in the chemistry cycle of 137 forest ecosystems. 138

Although there have been studies on the spatio-temporal variability of rainfall redistribution, most of these are limited to data of short-term monitoring over one-two years or several months (Liu et al., 2019; Ziegler et al., 2009; Carlyle-Moses, 2004; Marin et al., 2000). There are few studies exceeding several years experiments and focusing on forest structural changes and rainwater interception (Grunicke et al., 2020; Shinohara et al., 2015; Jackson, 2000). Long-term field monitoring studies are considered to be valuable to gain insight into the temporal dynamics of forest hydrological processes (Rodrigues et al., 2022; Sun et al., 2023; Levia and Frost, 2006). Such studies can also contribute to identify patterns and trends in rainfall redistribution, which is essential for predicting the long-term effect of water resource change on forest ecosystems.

Therefore, in this study, we focus on the changing characteristics of throughfall 150 and stemflow in a subtropical forest succession sequence (pine forest -> mixed forest -> 151 monsoon evergreen broadleaf forest), based on long-term monitoring. Specifically, the 152 153 objectives are to analyze: (1) the changes of water volume of throughfall and stemflow among the three forests, (2) the changes of water chemistry (TN, TP, K<sup>+</sup>) of rainfall, 154 throughfall and stemflow among the three forests. We hypothesize that: (1) both 155 throughfall and stemflow in the broadleaf forest were characterized with high 156 variability compared to mixed forest followed by pine forest, (2) chemistry flux of 157 throughfall and stemflow changed over time with in order broadleaf forest > mixed 158 forest > pine forest. We aim to assess the variability of forest hydrological processes 159 from a long-term perspective to help predict future dynamic trends of water resources 160 in subtropical forest ecosystems. 161

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#### 163 2. Materials and Methods

164 *2.1. Study site* 

165 This study was conducted at the Dinghushan Biosphere Reserve ( $23^{\circ}09' 21'' \text{ N} \sim$ 23°11′ 30″ N, 112°30′ 39″ E ~ 112°33′ 41″ E) located in Zhaoqing City, South China. 166 Dinghushan catchment consists of two streams both with 12 km length, which flow into 167 the West River (the main trunk of the Pearl River). According to the Köppen-Geiger 168 climate classification (Kottek et al., 2006), the study area belongs to tropical monsoon 169 climate (Cwa) with pronounced wet (April-September) and dry season (October-170 171 March). The average annual temperature is 20.9 °C, and the annual rainfall and evaporation are 1900 mm and 1115 mm, respectively. Dinghushan Biosphere Reserve 172 is covered with a complete horizontal succession series of three types of subtropical 173 174 forest, which is highly representative of the region (Zhou et al., 2011). Monsoon evergreen broadleaf forest (BF) is 400 years old with typical tree species including 175

Castanopsis chinensis (Spreng.) Hance, Schima superba Gardner & Champ., 176 Cryptocarya concinna Hance, etc. The mixed pine/broadleaf forest (MF) is a natural 177 succession with a coniferous broadleaf ratio of about 4:6, and 70-80 years old. The 178 main broadleaf tree species are Schima superba Gardner & Champ., Castanopsis 179 chinensis (Spreng.) Hance, and the coniferous species Pinus massoniana Lamb. The 180 pine forest (PF) planted before 1960 belongs to the primary succession community 181 where *Pinus massoniana Lamb* forms the only tree layer. The community composition 182 183 and biodiversity are shown in Table S1.

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# 185 2.2. Gross rainfall, throughfall and stemflow monitoring

186 Atmospheric rainfall data was collected at Dinghushan Automatic Meteorological Station from 2001–2022. Automatic meteorological systems were used to measure 187 atmospheric pressure (DPA501 gas-pressure meter), temperature (HMP45D sensor), 188 relative humidity (HMP45D sensor), rainfall (SM1-1 pluviometer), etc. Datalogger 189 190 (America Campbell, CR1000X) was used to control measurement sensors and store meteorological data. The resolution of data recording was  $\pm 0.2$  mm with a time interval 191 of 10 min. The raw data comprised annual rainfall amounts, as well as single rainfall 192 events with throughfall and stemflow measurements. 193

Throughfall and stemflow were collected in all three forest types and synchronously measured. Devices with cross-shaped collectors  $(1.25 \text{ m}^2)$  attached to reservoirs (1000 L) at the bottom were used to collect throughfall. Three throughfall devices were randomly installed in each forest field (Fig. S1).

Half-shell plastic tubes were installed around tree trunks attached to reservoirs 198 199 (1000 L) at the bottom to collect stemflow. The ratio of volume (mL) to canopy area (cm<sup>2</sup>) is the stemflow (mm). A total of 24 trees were selected to measure stemflow 200 201 volume (Table S2). In detail, four tree species were selected in the broadleaf forest, 202 including Acmena acuminatissima (Blume) Merr. et Perry (SF1), Cryptocarya chinensis (Hance) Hemsl. (SF2), Gironniera subaequalis Planch. (SF3), Schima 203 superba Gardn. et Champ. (SF4), with 3 repetitions respectively. Three tree species 204 were selected in the mixed forest, including *Castanea henryi* (Skam) Rehd. et Wils. 205 (SF5), Schima superba Gardn. et Champ. (SF6), Pinus massoniana Lamb. (SF7), with 206 207 3 repetitions respectively. In the pine forest, Pinus massoniana Lamb. (SF8) was selected as the monitoring subject with 3 repetitions. 208

# 210 2.3. Rainwater chemistry measurement

For the measurement of rainwater chemistry, rainwater samples were from 2000, 2010 and 2022 in Dinghushan area. The samples of open rainfall, throughfall and stemflow were manually collected for every one-month period, respectively. The samples of open rainfall and throughfall were collected with three repetitions, respectively and stemflow with four repetitions in the broadleaf forest, three repetitions in the mixed forest and three repetitions in the pine forest. In total, 792 rainwater samples (108 open rainfall, 324 throughfall and 360 stemflow) were collected.

Rainwater samples were defrosted and filtered through 0.45  $\mu$ m polypropylene membranes. Concentrations of total nitrogen (TN) and total phosphorus (TP) were measured using ultraviolet spectrophotometer (Lambda 25, Perkin-Elmer), and ion potassium (K<sup>+</sup>) was measured using an inductively coupled plasma optical emission spectrometer (Optima 2000, Perkin-Elmer), respectively. The origin data of TN, TP and K<sup>+</sup> were processed into annual flux and monthly values by weighted average method,

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$$C = \frac{\Sigma c_i * v_i}{\Sigma v_i} \tag{1}$$

where  $C_i$  and  $V_i$  are the concentrations of ions (mg L<sup>-1</sup>) and water sample volume (L) in each rainfall event, respectively.

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# 228 2.4. Other measurement and statistical analysis

229 In the forests, plant density and canopy structure have been measured every five years since 2000. 25 plots of 20 m  $\times$  20 m (A1-A25 plots) were built on a plot of 1 hm<sup>2</sup> 230 to survey tree density (Fig. S1). Then, 25 plots of 5 m  $\times$  5 m (B1-B25 plots) were 231 randomly set on the A1-A25 plots to survey shrub density. Finally, 25 plots of  $1 \text{ m} \times 1$ 232 233 m (C1-C25 plots) were randomly set on the B1-B25 plots to survey herb density. The percentage of the surface area covered by plants to the total plot area is termed canopy 234 coverage (%). 25 observation plots  $(1 \text{ m} \times 1 \text{ m})$  were selected in the 1 hm<sup>2</sup> area of each 235 forest type. LAI (Leaf area index) was measured using a LAI-2200 plant canopy 236 analyzer with 90° view caps (Li-Cor Inc., USA). 10 observation points (distance about 237 10 m) were selected in the 1 hm<sup>2</sup> area of each forest type with 5 replications. Growth 238 indicators of the selected trees have been recorded: tree height (m), diameter at breast 239 height (DBH, cm), and crown area (CA, m<sup>2</sup>). Tree height was measured using laser 240 range finder. Tape measure was used to measure the diameter of trees at a height of 1.3 241

m, namely DBH. CA: the laser rangefinder was used to measure the maximum diameter
at the edge of the canopy, with multiple measurements at different points to ensure
accuracy.

The differences in throughfall and stemflow among different forests were assessed using analysis of variance (ANOVA), followed by a Tukey test for multiple comparisons between means. Mann-Kendall (MK) test was used to analyze the variation trend of annual rainfall. All statistical procedures were conducted with  $\alpha =$ 0.05 threshold for significance, in the IBM SPSS statistics 22.0 software (IBM Inc.).

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# **3. Results**

# 252 *3.1. Rainfall and temperature characteristics*

Based on the 22 years rainfall dataset from the Dinghushan area, annual gross 253 254 rainfall ranged between 1370.0 and 2361.1 mm (Fig. 1). 78.0% of gross rainfall appeared in the wet season (April–September). Result of M-K test showed that rainfall 255 was in a significantly decreasing trend from 2001–2007 (UF < 0, P < 0.05), and shifted 256 into a significantly increasing trend from 2012–2022 (UF > 0, P < 0.05). Moreover, 257 2008 and 2011 (the intersection of UF and UB) were the mutation time of rainfall trend 258 (P < 0.05). Anomaly were revealed in the temporal variability (coefficient of variation, 259 CV of 16.6%) in annual rainfall (Fig. 1a). Anomaly varied at -426.4-476.8 mm and -260 258.0-471.4 mm in the wet season and dry season, respectively. By comparison, dry 261 season experienced greater variation with CV of 40.4% than wet season with CV of 262 21.7%. Besides, annual raining days obviously tended to decrease over time from 2012 263 to 2021(Fig. 1b). Based on five rainfall classifications, it was shown among 22 years 264 that rainfall <10 mm account for about 68.5% of total raining days (2856), while 265 rainfall >50 mm account for about 4.9%. Besides, annual temperature changed between 266 21.8 °C and 23.3 °C over 22 years. Result of M-K test showed statistically significant 267 rising trend for temperature for 8 years out of 22 years (UF > 0, P < 0.05). Moreover, 268 269 2005 and 2013–2014 were the mutation time of temperature trend (P < 0.05).

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# 271 *3.2 Variability of throughfall*

Rainfall redistribution (throughfall and stemflow) among the three forests all experienced differing magnitude during 22 years (Fig. 2). Annual throughfall were concentrated between 954.2 mm and 2192.6 mm. Result of M-K test showed that

throughfall was in a significantly decreasing trend at first (UF < 0, P < 0.05) and then 275 shifted into an significantly increasing trend (UF > 0, P < 0.05) from 2001–2022, 276 similar with the trend of open rainfall. Differently, the mutation of throughfall trend 277 occurred in 2008, 2011 and 2021 in the broadleaf forest, 2008 and 2011 in the mixed 278 forest, 2006, 2008, 2011 and 2021 in the pine forest (P < 0.05). For throughfall ratio, 279 it varied significantly both at the event and interannual scales (Fig. 3a, b and c). The 280 median of annual throughfall ratio in the broadleaf forest varied between 60% and 120% 281 with CV of 16.4% from 2001–2022. The median of throughfall ratio in the mixed pine 282 283 and broadleaf forest varied between 80% and 110% with a CV of 9.7%. The median of throughfall ratio in the pine forest varied between 59% and 110% with a CV of 11.8%. 284 Therefore, throughfall ratio was characterized by a relatively low variability over 285 annual-time scale (CV < 20%). Besides, some differences of throughfall ratio were 286 found among the three forest types based on rainfall classifications (Fig. 3d). For 287 rainfall events <10 mm, throughfall ratio range in the broadleaf forest was 30%-70%, 288 while in the other two forest types it was 15%-85%. The mean value of throughfall 289 290 ratio was relatively small in the broadleaf forest (53.9%), though no significant difference among the three forests (P > 0.05) were detected. For rainfall events <50 mm, 291 292 no significant difference of throughfall ratio among the three forest types was found (P >0.05). However, the median values of throughfall ratio in the pine forest (90.0%) and 293 294 the mixed forest (89.4%) were both significantly larger than that in the broadleaf forest (83.7%) for rainfall events >50 mm. 295

CV values of throughfall based on all the rainfall event classifications were drawn 296 in the Fig. 4a. Results showed that median CV of throughfall in the pine forest (15.2%) 297 was lower than that for the broadleaf forest (21.7%) and for the mixed forest (26.3%)298 for rainfall events <10 mm. For rainfall events >10 mm, small differences of median 299 300 CV among the three forest types were shown. Meanwhile, CV values decreased with the increasing rainfall events, eventually falling to 3.5%-4.3%. Besides, CV values of 301 throughfall based on interannual scale were drawn in the Fig. 5a, b and c. Annual CV 302 values among different forest types showed different fluctuations over time. The 303 304 medians of CV in annual, wet and dry seasons presented different order in different years. According to linear fitting, significant negative correlations were found in the 305 median of  $CV_{TF}$  in the mixed forest over time (r = 0.63, P < 0.01). In addition, fitting 306 result of in total 740 rainfall events in 22 years showed that CV values of throughfall 307 significantly decreased with increasing gross rainfall (Fig. S2). 308

# 310 *3.3 Variability of stemflow*

Annual stemflow was concentrated between 9.0 and 119.7 mm over 22 years (Fig. 311 2). More stemflow were collected in the broadleaf forest and mixed forest than in the 312 pine forest. Result of M-K test showed that stemflow of both broadleaf forest and mixed 313 forest were in a decreasing trend at first (UF < 0, P < 0.05) and then shifted into an 314 increasing trend (UF > 0, P < 0.05), different from continuously increasing trend of 315 pine forest (UF > 0). The mutation of stemflow trend occurred in 2008 in the broadleaf 316 317 forest, 2011 in the mixed forest, 2006, 2012 and 2015 in the pine forest (P < 0.05). Stemflow ratio changed significantly both at the event and interannual scales (Fig. 3a, 318 b and c). Among 22 years, the median of annual stemflow ratio in the broadleaf forest 319 varied between 1.3% and 5.4% with a CV of 56.2%. The stemflow ratio of mixed forest 320 varied between 1.5% and 4.4% with a CV of 38.6%. In the pine forest, it varied between 321 0.3% and 1% with a CV of 50.9\%. This indicated that the stemflow ratio was 322 characterized by an extremely high variability over time. Same to the seasonal 323 throughfall ratio, the medians of stemflow ratio in annual, wet and dry seasons 324 presented different orders in different years. Besides, the stemflow ratio significantly 325 326 changed among tree species and among rainfall classifications (Fig. 3e). By comparison, stemflow ratios of the SF1 and SF2 trees in the broadleaf forest were both higher in all 327 the tree species for the rainfall events <50 mm. However, for strong events (>50 mm), 328 the stemflow ratio of the SF5 tree in the mixed forest was highest for all tree species, 329 followed by the trees in the broadleaf forest. For all the rainfall events, the stemflow 330 ratio of SF7 in the mixed forest and SF8 in the pine forest were both lower than that for 331 other tree species. 332

CV values of stemflow based on rainfall event classifications were drawn in the 333 Fig. 4b. By comparison, stemflow varied more than those of throughfall across rainfall 334 events, with CV<sub>SF</sub> values of 25%–130%. Median CV of stemflow in the pine forest was 335 always lower (45%–68%) than that for the other two forest types (56%–120%). CV 336 values of stemflow based on interannual scale changed over time among different forest 337 types (Fig. 5d, e and f). The medians of  $CV_{SF}$  in annual, wet and dry seasons presented 338 different order in different years. By comparison,  $CV_{SF}$  was always greater than  $CV_{TF}$ , 339 and interannual fluctuation of CV<sub>SF</sub> was also stronger than CV<sub>TF</sub>. According to linear 340 fitting, significant negative correlations were found in the median of  $CV_{SF}$  in the 341 broadleaf forest over time (r = 0.73, P < 0.001)). In addition, fitting result of in total 342

740 rainfall events in 22 years showed that *CV* values of stemflow both significantly
decreased with increasing gross rainfall (Fig. S2).

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#### 346 *3.4. Rainwater chemistry*

Rainwater (open rainfall, throughfall and stemflow) chemical properties (TN, TP 347 and  $K^+$  concentration) were measured in the three forest types, respectively. All of TN, 348 TP and  $K^+$  values presented in order stemflow > throughfall > open rainfall (Fig. 6a, b 349 and c). However, changes of TN, TP and  $K^+$  were different for the three forest types 350 among 2000, 2010 and 2022. For instance, in 2000 and 2010, TN values of throughfall 351 and stemflow decreased for both in order of pine forest > mixed forest > broadleaf forest, 352 353 while no such result could be confirmed in 2022. Similarly, TP values of throughfall in broadleaf forest was 1.3 times higher than that in pine forest in 2022, while TP values 354 in pine forest was 6.8 times than that in broadleaf forest in 2000. K<sup>+</sup> values of stemflow 355 in 2010 (6.76 mg L<sup>-1</sup>) and 2022 (6.22 mg L<sup>-1</sup>) were higher for broadleaf forest than 356 those for pine forest (3.76 mg L<sup>-1</sup> and 2.46 mg L<sup>-1</sup>), which was different from that in 357 2022. 358

TN, TP and K<sup>+</sup> fluxes of stemflow were < 10 kg ha<sup>-1</sup> a<sup>-1</sup>, 0.2 kg ha<sup>-1</sup> a<sup>-1</sup>, 6 kg ha<sup>-1</sup> 359  $a^{-1}$ , respectively, all lower than those of throughfall and open rainfall (Fig. 7d, e and f). 360 In the 2000, 2010 and 2022, TN flux (39.4–87.4 kg ha<sup>-1</sup> a<sup>-1</sup>) was 1.2–1.8 times greater 361 than that of open rainfall, 3.3–28.0 times greater than that of stemflow. TP flux (1.1– 362 2.7 kg ha<sup>-1</sup> a<sup>-1</sup>) was 1.0–2.3 times greater than that of open rainfall, 8.7–31.4 times 363 greater than that of stemflow.  $K^+$  flux (21.5–59.2 kg ha<sup>-1</sup> a<sup>-1</sup>) was 2.2–8.1 times greater 364 than that of open rainfall, 2.2–26.8 times greater than that of stemflow. In addition, TN, 365 TP and K<sup>+</sup> fluxes of stemflow increased with succession from primary to climax, 366 namely pine forest<mixed forest<br/>broadleaf forest. Different from this, differences in 367 chemistry fluxes of throughfall was not found among different forests, neither among 368 different periods. 369

Besides, monthly chemistry concentrations in rainfall, throughfall and stemflow showed distinct changes (Fig. 7). Monthly TN, TP and K<sup>+</sup> concentrations of rainfall were always lower than those of stemflow for all trees. Monthly TN, TP and K<sup>+</sup> of stemflow in the dry season were generally higher than in the wet season. High monthly TN concentrations of stemflow with SF6 of mixed forest and SF8 of pine forest were found, especially in dry season with maximum TN concentrations of 27.59 mg L<sup>-1</sup> at 376 SF6 and 19.94 mg L<sup>-1</sup> at SF8, respectively. Differently, high monthly  $K^+$  concentration 377 of stemflow at SF4 in broadleaf forest was found, with in dry season maximum  $K^+$ 378 concentration of 25.17 mg L<sup>-1</sup>.

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### 380 4. Discussion

# 381 *4.1. Open rainfall partitioned to throughfall and stemflow*

Studies in forests have confirmed that throughfall volume increased with 382 increasing gross rainfall at event scale, accounting for 60%-80% of gross rainfall (Ji et 383 al., 2023; André et al., 2011; Carlyle-Moses, 2004). Throughfall ratio changed over time 384 and showed different fluctuations among different forests (Fig. 3). During light rainfall 385 386 events with rainfall amounts <10 mm, a low proportion of raindrops would reach the ground as throughfall, as the tree canopy intercepts almost all the incoming raindrops. 387 Specifically, high canopy coverage in broadleaf forest can reinforce raindrop intercept 388 (Brasil et al., 2018; Ponette-González et al., 2010), consequently generating lower 389 390 throughfall ratio than those in the mixed forest and pine forest (Fig. 3). During moderate rainfall events (10–50 mm), given that the intercept effect of the wetting tree canopy 391 was weakened (Shinohara et al., 2015), throughfall ratio was in a high and steady state. 392 As the gross rainfall increases further (>50 mm), significant differences of throughfall 393 ratio were found among the three forests. Throughfall ratio was significantly lower in 394 395 the broadleaf forest than those in the other two forests. Likewise, such differences due to rainfall event class also appeared in other forest studies with stands such as beech, 396 pine in monocultures and mixed pine-beech (Blume et al., 2022). Influenced by forest 397 stand characteristics, throughfall therefore indicated different forest water budget. 398

Stemflow of forests were variably controlled by tree species, on average 399 400 accounting for about <10% of gross rainfall, even lower (<1%) (Sun et al., 2018; André et al., 2008; Crockford and Richardson, 1990). In our study site, the lowest stemflow 401 (<1%) was collected in the pine forest, though weakly increasing with rainfall 402 classifications (Fig. 3). Stemflow ratio in broadleaf forest was maintained at 5%-10% 403 without the effect of rainfall amount seemingly. In detail, stemflow ratio of pine forest 404 405 (SF8) was significantly lower than those of broadleaf forest (SF1~4). And in the mixed forest, broad-leaved trees (SF5 and SF 6) have larger stemflow than pine tree (SF7). 406 However, for some rainfall events, extraordinary low proportion of stemflow in the 407 408 broadleaf forest and extraordinary high proportion in the pine forest were caught. This 409 implied the key role of rainfall conditions (e.g., intensity, duration) and tree species with tree traits (e.g., branch angle), consistent with reported studies e.g., in evergreen forest 410 (Chen et al., 2019; Bruijnzeel et al., 2011) and pine forest (Pinos et al., 2021; Crockford 411 and Richardson, 1990). Moreover, ANOVA showed significant differences of stemflow 412 ratio among tree species and rainfall classifications (P < 0.001) (Table 1). This indicated 413 that rainfall and tree species simultaneously affect stemflow. Branch inclination angle, 414 canopy cover, tree height and DBH of tree species proved to be key factors in stemflow 415 yield (Levia et al., 2015). 416

417 Throughfall and stemflow were generally enriched in chemical concentration compared with open rainfall due to leachable canopy/stem ion pools (Jiang et al., 2021; 418 Van Stan et al., 2017; Zimmermann et al., 2007). In our study, the concentration of K<sup>+</sup> 419 in stemflow was 16 times higher than that in open rainfall and in throughfall reached 420 up to 11 times higher than open rainfall (Fig. 6). Similar results were also found in 421 artificial plantation (Acacia mangium and Dimocarpus longan) of South China (Shen 422 et al., 2013), in Oriental beech (Fagus orientalis Lipsky) trees in Northern Iran (Moslehi 423 et al., 2019), indicating strong  $K^+$  leaching from canopy. Even so, throughfall was 424 generally characterized with high fluxes compared to open rainfall followed by 425 426 stemflow, it thus is the largest contributor to wet deposition. Meanwhile, TN flux of throughfall was greatest in the pine forest in 2010, TP flux of throughfall was greatest 427 in the broadleaf forest in 2000, and K<sup>+</sup> flux of throughfall was greatest in the mixed 428 forest in 2010. It should be noted that the differences of rainwater chemistry shifted 429 430 over time among the three forests. Accordingly, throughfall and stemflow via canopy and stem input soil is a significant contributor, and its long-term effect on ecosystems 431 432 needs more attention (Fan et al., 2021). After all, atmospheric wet deposition provides nutrient requirement for ecosystems, but also imposes a considerable burden on the 433 434 ecosystems in general. For instance, N enrichment and P limitation have proven to have different effect on soil carbon sequestration, microbial community composition and 435 forest productivity, especially in tropical and subtropical forest ecosystems with highly 436 weathered soils (Zheng et al., 2022; Li et al., 2016; Huang et al., 2012). Besides, 437 438 throughfall and stemflow was mainly characterized by low chemical concentrations in the wet season and high concentrations in the dry season. Primary reasons for seasonal 439 rainwater chemistry may be attributable to moisture source associated with frontal 440 weather systems and gradually depleting effect with increasing rainfall amount 441 (Dunkerley, 2014; Germer et al., 2007). The present study in subtropical forests and 442

443 previous studies in tropical forests and European temperate forests all exhibited variable 444 rainwater chemistry in throughfall and stemflow, both spatially and temporally 445 (Zimmermann et al., 2007; Staelens et al., 2006; Seiler and Matzner, 1995). In fact, the 446 chemical concentration of rainfall redistribution was also affected profoundly by 447 canopy and stem parameters of tree species (Tonello et al., 2021b; Chen et al., 2019). 448 In our study, some differences of TN, TP and K<sup>+</sup> were also found among SF1~SF8 due 449 to tree-species specific effect (Legout et al., 2016; De Schrijver et al., 2007).

450

#### 451 *4.2. Long-term changes of rainfall in forests*

Rainfall regimes induce divergent spatially hydrological changes (Wu et al., 2024). 452 Likewise, our study found that throughfall of forests experienced a decreasing followed 453 by an increasing from 2001–2022, similar with the trend of open rainfall, and stemflow 454 showed characteristic trends in different forests especially in the pine forest (Fig. 1). 455 This suggested that the complexity of forest structure and rainfall amount and their 456 change exacerbated the spatio-temporal variability of throughfall and stemflow. Firstly, 457 interannual variability of forest structure (e.g., canopy coverage, leaf area index) and 458 tree parameters (e.g., height, DBH and CA) made throughfall and stemflow distribution 459 460 uncertain (Yue et al., 2021). From 2001 to 2022, changes in forest structure were confirmed in all three forests, such as changes in plant density, canopy coverage and 461 LAI (Fig. 8). Throughfall ratio and stemflow ratio in the succession forest systems all 462 varied over time accordingly. Similarly, driven by forest structure (e.g., tree density, 463 species dominance), a six-year dataset from the Brazilian Atlantic Forest showed that 464 the spatial variability of throughfall over time was less stable (Rodrigues et al., 2022). 465 Besides, the variation of stemflow  $(CV_{SF})$  was obviously larger than that of throughfall 466  $(CV_{\rm TF})$  (Fig. 4), which probably was attributed to the differences of tree species in 467 468 stemflow (Fig. 3). For a forest succession, a 17 years' study showed that the shift from monoculture Japanese red pine to mixture of red pine, evergreen oak and theaceous tree 469 made stemflow significantly increasing (Iida et al., 2005). Likewise, for the forest 470 succession in Dinghushan area, stemflow ratio in broadleaf forest and mixed forest were 471 472 both higher than that in tree-monospecific pine forest. High plant density (tree and shrub) and LAI in broadleaf forest and mixed forest conduce to rainwater interception of multi-473 canopy trees through more leaves and angled branches, which potentially enhanced 474 stemflow (Fig. 8). Indeed, some differences of rainfall redistribution appeared in multi-475 layered vegetative structure. An experiment on vegetation communities with a complex 476

477 multi-layered structure found that interception loss from shrubs was two-times higher than from trees, and smaller trees generated stemflow more efficiently than the higher 478 ones (Exler and Moore, 2022). Based on the 22 years' data from forest community 479 survey in our study site, forest canopy parameters (e.g., coverage and LAI) of trees and 480 shrubs showed variation over time from 2001 to 2022 (Fig. 8). In the broadleaf forest, 481 482 plant density of trees and canopy coverage of shrubs showed a slight increment compared to the other two forests, though LAI was decreasing. During this period, 483 484 interannual throughfall ratio and stemflow ratio showed significantly change over time 485 (Fig. 3), implying the role of interannual variation of forest structure in rainfall 486 redistribution process.

Secondly, ongoing rainfall changes with different magnitude favor the different 487 levels of rainfall redistribution over time (Lian et al., 2022). At event scale, throughfall 488 and stemflow proportions of forests were both low with rainfall events <10 mm. The 489 490 variations of throughfall and stemflow were both larger for gross rainfall <10 mm than 491 events >10 mm. Rainfall threshold associated with the canopy interception capacity had 492 impact on throughfall and stemflow generation (Zabret et al., 2018; André et al., 2008; Durocher, 1990). After the raindrop capacity of the canopy reached its peak, throughfall 493 494 and stemflow were documented to match the gross rainfall. Therefore, relatively low proportions and high spatial variability appeared before rainfall threshold, and after that, 495 496 relatively high proportions and low variability until a stable level were observed in the three forests. Moreover, at interannual scale, the raining days in different magnitudes 497 498 presented obvious fluctuation over 22 years (Fig. 1). This fluctuation of raining days 499 and its magnitude distribution potentially regulated the long-term changes of open 500 rainfall partitioned to interception loss, throughfall and stemflow. Consequently, 501 throughfall and stemflow, influenced by the comprehensive effect of rainfall regimes 502 and forest structures, presented spatiotemporal variability at different level (Fig. 3-6). From a long-term perspective, changing in rainfall redistribution potentially makes 503 forest water and biogeochemistry budget more complex. Further knowledge of the 504 long-term accumulative effect of rainfall redistribution on forest water and chemistry 505 506 (e.g., soil and plant) is needed in the future.

507 Throughfall and stemflow are part of rainfall and are key player in the water cycle 508 process. Based on the connection of the water cycle to precipitation and temperature 509 and under the background of climate change, frequency of extreme events (heavy 510 rainfall, droughts) needs to be anticipated in the effect on rainfall redistribution and

solute transport within forests, which in turn may affect the water cycle and 511 biogeochemical cycles (Blume et al., 2022). In this study, it should be noted that the 512 2008 rainfall data can be used as an example under extreme event. In 2008, extreme 513 weather events occurred in South China. Freezing events occurred in the dry season, 514 continuous heavy rain and typhoon events occurred in the wet season. Gross rainfall 515 516 was larger than other years, with the annual rainfall of 2361.1 mm (22-year average annual rainfall of 1848.6 mm) (Fig. 1). At the same time, a total of 26 throughfall events 517 were collected in 2008. According to the M-K test, the throughfall and stemflow trend 518 519 of different forests presented different degree of disturbance under the background of mutation of open rainfall (Fig. 2). In this process, the driving effect of forest structure 520 and rainfall on throughfall and stemflow mutation is synchronous. More data and 521 modeling are needed to support the relevant study about effect of climate change on 522 rainfall redistribution in the future. 523

524

# 525 **5. Conclusion**

The current study investigated long-term changing characteristic of rainfall 526 redistribution along a subtropical forest succession sequence with: pine forest (PF), 527 528 mixed pine and broadleaf forest (MF) and monsoon evergreen broadleaf forest (BF). Firstly, in the valid 740 rainfall events throughfall ratio showed in order BF < MF < PF, 529 530 and stemflow ratio showed in order BF > MF > PF. The variation of stemflow was higher (CV >50%) than that of throughfall (CV <25%). Secondly, throughfall and 531 532 stemflow of forests experienced a decreasing followed by an increasing from 2001-2022 (except stemflow of the pine forest), similar with the trend of open rainfall. Driven 533 534 by rainfall and forest factors, interannual variability of both throughfall and stemflow in the broadleaf forest were greater than those in the mixed forest and pine forest, which 535 was different from that of annual open rainfall. 536

For rainwater chemistry, differences of the clement flux in throughfall and 537 stemflow among the three forest types were confirmed based on data from 2001, 2010 538 and 2022. On average, TN and TP fluxes of throughfall presented in order BF < MF <539 PF, while  $K^+$  flux of throughfall presented in order BF > MF > PF. Over time, rainwater 540 chemical concentrations were lower in the wet season than that in the dry season. Given 541 the smaller proportion of open rainfall, stemflow chemical fluxes varied less among 542 forest types and/or over time, though tree species exactly contribute to differences in 543 stemflow chemistry. Nevertheless, its funnel effect on soil and plant over time still 544

545 deserves more attention in the future.

The above results indicate that the water volume and chemistry in rainfall redistribution process under forest represented not exactly the same trend as open rainfall over time, and throughfall and stemflow depend on the effect of rainfall and forest factors. This study thus provided insight into the rainfall redistribution process by linking the long-term change of rainfall pattern with a subtropical forest succession sequence.

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- 553

554 *Code and data availability*. The data used to derive to the conclusions of the present 555 study are freely accessible. All the data were obtained from the CNERN dataset 556 (http://dhf.cern.ac.cn/meta/metaData).

557

*Author contributions*. WJZ: conceptualization, investigation, data analysis, writing, visualization. TS and SS: reviewing, supervision. QMZ and CGW: resources, data curation, WLH: reviewing, JXL and XX: reviewing, funding acquisition, supervision

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562 *Competing interests.* The authors declare that they have no conflict of interest.

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#### Tables 833

#### 834

Table 1 Correlations between throughfall and stemflow and rainfall and forest factors 835

G	ross rainfall	DBH	CA	Height	LAI	
Throughfall 0.	72***				-0.58**	
stemflow 0.	77***	0.65*	-0.75**	$0.54^{**}$	-0.55**	
DBH: diameter at bi	: at breast height; CA: crown area; LAI: leaf area index. $*P < 0.05$ , $**P < 0.01$ , $**$					
0.001						
Table 2         Analysis	of variance	(ANOVA)	for throughfall	and stemflow a	ffected by ra	
classifications and tree species						
Summary of ANO	VA T	Throughfall			Stemflow	
Rainfall classificat	(R) <	0.05	Rainfall	classification (R)	< 0.001	
Forest type (F)		0.001	Tree spe	cies (T)	< 0.001	
$\mathbf{R} \times \mathbf{F}$	0	.861	$R \times T$		< 0.001	

 $\alpha = 0.05$ 846

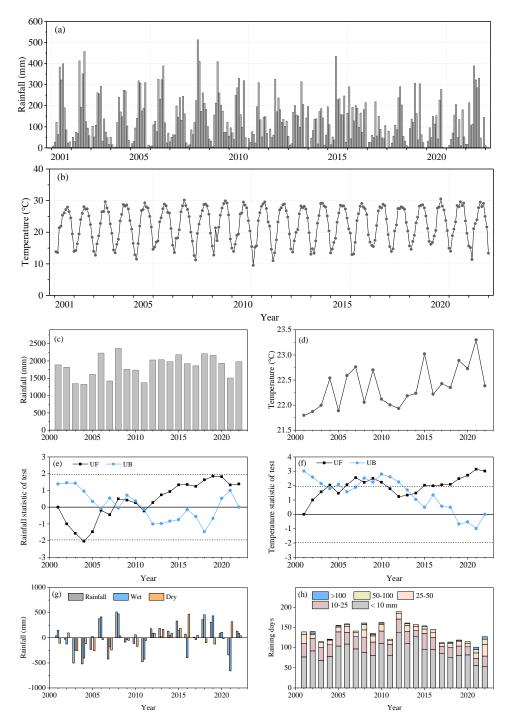
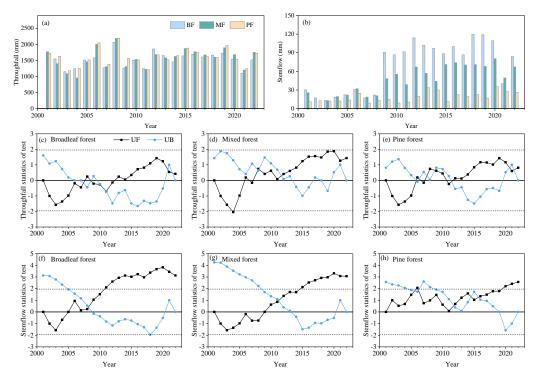


Fig. 1 (a) and (b) rainfall and temperature in Dinghushan Biosphere Reserve in Southern China from 2001–2022, (c) and (d) annual rainfall and temperature, (e) and (f) rainfall and temperature statistic of Mann-Kendall test, respectively. (g) Anomaly of annual rainfall from 2001–2022, (h) annual raining days in five classifications. UF (Unadjusted Forward) > 0 indicate a continuous increasing trend (P < 0.05). The intersection points of UF and UB (Unadjusted Backward) is the mutation time point. Within the confidence interval [-1.96, 1.96], the variable presents a significantly mutation growth state at this time point (P < 0.05).



859Fig. 2 (a) and (b) Annual throughfall and stemflow in the broadleaf forest (BF), mixed pine and860broadleaf forest (MF) and pine forest (PF) from 2001–2022, respectively, (c) ~ (h) rainfall and861stemflow statistic of Mann-Kendall test, respectively. UF (Unadjusted Forward) > 0 indicate a862continuous increasing trend (P < 0.05). The intersection points of UF and UB (Unadjusted863Backward) is the mutation time point. Within the confidence interval [-1.96, 1.96], the variable864presents a significantly mutation growth state at this time point (P < 0.05).

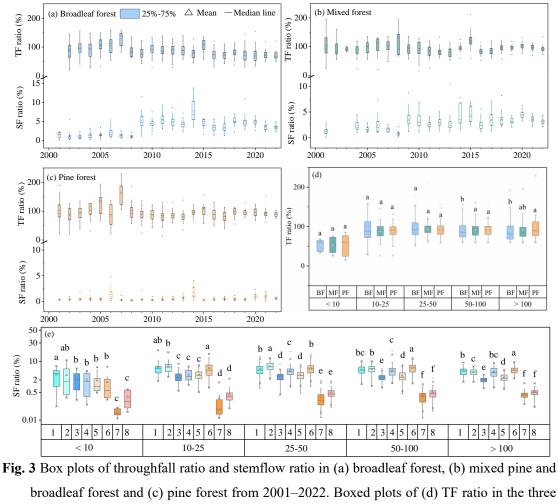
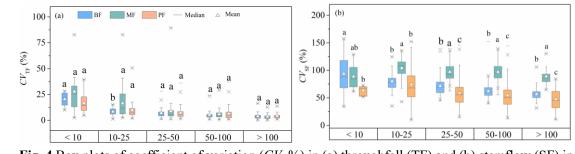
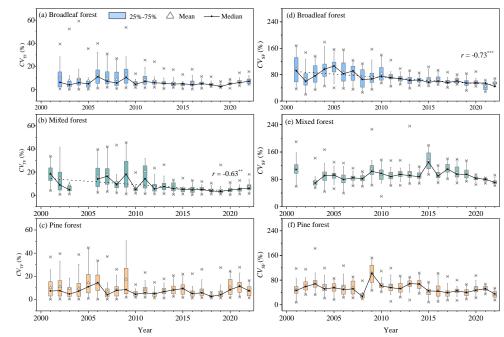


Fig. 3 Box plots of throughfall ratio and stemflow ratio in (a) broadleaf forest, (b) mixed pine and
broadleaf forest and (c) pine forest from 2001–2022. Boxed plots of (d) TF ratio in the three
forests and (e) SF ratio for eight plant species based on the rainfall classifications (broadleaf
forest: *Acmena acuminatissima* (Blume) Merr. et Perry (SF1), *Cryptocarya chinensis* (Hance)
Hemsl. (SF2), *Gironniera subaequalis* Planch. (SF3), *Schima superba* Gardn. et Champ. (SF4);
mixed forest: *Castanea henryi* (Skam) Rehd. et Wils. (SF5), *Schima superba* Gardn. et Champ.
(SF6), *Pinus massoniana* Lamb. (SF7); pine forest: *Pinus massoniana* Lamb. (SF8). Different
letters indicate a significant difference at *P* < 0.05</li>



876Fig. 4 Box plots of coefficient of variation (CV, %) in (a) throughfall (TF) and (b) stemflow (SF) in877broadleaf forest (BF), mixed pine and broadleaf forest (MF) and pine forest (PF) based on the878rainfall classifications. Different letters indicate a significant difference at P < 0.05



881Fig. 5 Box plots of coefficient of variation (CV, %) in (a, b, and c) throughfall (TF) and (d, e, and f)882stemflow (SF) in the three forests from 2001 to 2022. Medians of annual CV were fitted. r:883Pearson coefficient of correlation; \*: P < 0.05, \*\*: P < 0.01, \*\*\*: P < 0.001

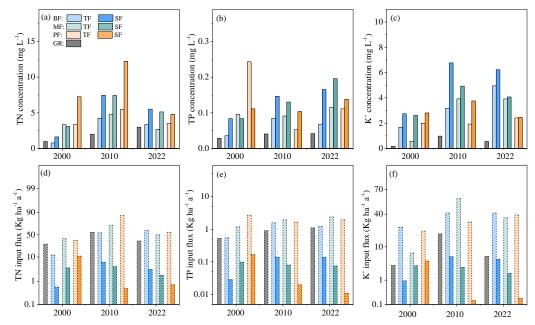
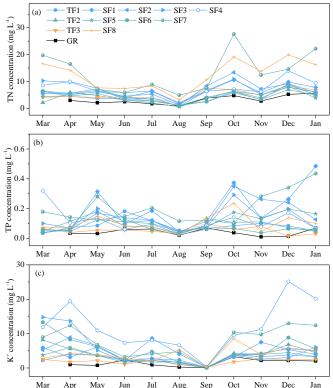


Fig. 6 Concentrations and fluxes of TN, TP and K<sup>+</sup> of gross rainfall (GR), throughfall (TF) and
stemflow (SF) in the broadleaf forest (BF), mixed forest (MF) and pine forest (PF) in 2000,
2010 and 2022, respectively.



Kig. 7 Monthly concentrations of (a) TN, (b) TP and (c) K<sup>+</sup> of throughfall (TF1) and stemflow (SF1, SF2, SF3 and SF4) in the broadleaf forest, throughfall (TF2) and stemflow (SF5, SF6 and SF7)

891 in the mixed forest, throughfall (TF3) and stemflow (SF8) in the pine forest.

