1    # Using Phosphate Fertilizer to Reduce Emitter Clogging of Drip

2    # Fertigation Systems with High Salinity Water

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9    **ABSTRACT**: Phosphorous (P) fertigation with high salinity water (HSW) drip irrigation would be an

10   effective measure to relieve soil and water pollution caused by the excessive application of P fertilizer, and

11   achieve synergistic saving of both limited fresh water and non-renewable P resources. However, the

12   emitter clogging issues of drip fertigation systems seriously restricts the utilization of this technology. This

13   study proposes an approach to reduce emitter clogging in HSW drip fertigation systems by choosing the

14   appropriate type and concentration of P fertilizer. The effects of two new types of P fertilizers (ammonium

15   polyphosphate, APP; urea phosphate, UP), and a traditional P fertilizer (monopotassium phosphate, MKP),

16   were assessed at three fertilization concentrations (0, 0.15, and 0.30 g/L) on the clogging behavior of four

17   types flat emitters. The results indicated that the application of MKP aggravated the clogging of emitters in

18   comparison with non-fertilization. While the addition of two new types of P fertilizers (APP and UP)

19   effectively alleviated emitters clogging (the irrigation uniformity of systems increased by 26.2% - 74.6%)

20   by inhibiting the formation of carbonate, although precipitation of phosphate, silicate, and quartz increased.

21   Moreover, under the equal application amount of P fertilizer, UP and APP were more effective in relieving

22   clogged when applied at a low-concentration with long-term running and high-concentration with

23   short-term running mode. The results could pave a way for reducing the pollution in agricultural

24   production and conserving freshwater and non-renewable P resources.

25   **Keywords**: brackish water; fertigation; drip irrigation; emitter discharge
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# 1. Introduction

Effective irrigation and fertilization managements in agricultural production are the most fundamental supports to ensure global food security and sustainable development of human society (Qin et al., 2019; Yu, et al., 2019). Over the past decades, the scale and intensity of agricultural production have been expanded rapidly. At present, agricultural irrigation accounts for 69% of global freshwater consumption (FAO, 2016), and nearly 1/5 of the world's population is living in water-scarce areas. Meanwhile, fertilizers are often excessively inputted in agricultural production due to its low utilization efficiency, which caused a serious agricultural non-point source pollution and water eutrophication (Vaneeckhaute et al., 2016; Chien, et al., 2009). In particular, the seasonal utilization efficiency of P always less than 25% (Wang et al., 2015) because P is easily immobilized by soil (Roberts, 2015). Moreover, the raw material of P fertilizer, e.g., phosphate rock as a non-renewable resource, is expected to be exhausted within 50 years based on current mining intensity (Natasha, 2009). Therefore, the scarcity of freshwater and P resources associated with water and soil pollution, and food security has become one of the important challenges in the 21st century (Li et al., 2019a; Scholz and Wellmer, 2015). Thus, there is a need for reducing the consumption of freshwater and P fertilizer and increasing their utilization efficiency in agricultural production.

The utilization of widespread high salinity water (HSW) may offer an effective measure to cope with water scarcity. However, irrigation with HSW can easily cause salt stress in the crop root zone, inhibiting its applications in agricultural irrigation (Wei et al., 2019). Drip irrigation is considered the most reliable and effective ways to utilize HSW (Karlberg et al., 2007), as salts usually accumulate at the edge of the wetting front, thus reducing the salt stress in the root zone (Kang et al., 2010). On the other hand, compared with the traditional application of P fertilizer (i.e. directly broadcast P into the soil), the application of dissolved P fertilizer through drip irrigation system can greatly increase the movement of P

49    and reduce the adsorption by soil, thereby improving the utilization efficiency of P fertilizer (Ayars et al.,

50    2017). Therefore, drip fertigation technology (i.e. integration of water and fertilizer) is expected to be a

51    promising way to cope with the soil and water pollution caused by the excessive input of P fertilizer and

52    save limited freshwater and P resources.

53        However, drip fertigation with HSW is currently not popular. The main reason is that the channel of

54    emitter in a drip irrigation system is very narrow (0.5 - 1.2 mm) (Goyal and Mansour., 2015), and many

55    salt ions in HSW can easily recombine and precipitate inside the channel, which induces emitter clogging

56    (Zhangzhong et al., 2016). Furthermore, the emitters clogging becomes more complex with addition of P

57    fertilizer. Zhou et al. (2019) found that the application of MKP greatly increased emitter clogging. Overall,

58    emitter clogging has become the main obstacle to the integrated application of drip fertigation with HSW

59    and P fertilizer, which would directly reduce the irrigation and fertilization uniformity in drip irrigation

60    systems, and greatly reduce the replacement cycle of drip lines of irrigation system. The appropriate

61    application mode of P fertilizer could relieve the clogging issue. Yang et al. (2019) observed that the rate of

62    drip emitters clogging obviously differed with different P fertilizers. Li et al. (2015) reported that the

63    concentration of fertilizer affected the clogging of drip emitters. In recent years, two new types of

64    phosphate fertilizers (UP and APP) have been identified might could alleviate emitter clogging. Every

65    kilogram of dissolved UP produces 6.3 moles of $H^+$, which makes it a high concentration acidifier (Goyal,

66    2015) to avoid the mineral precipitation (e.g. calcium carbonate). Polyphosphate does not easily react with

67    calcium and magnesium ions in water under specific conditions, and it is often used as scale inhibitor (Noy

68    and Yoles, 1979). However, effect of the two new phosphate fertilizers on the clogging of emitters in drip

69    irrigation system using HSW have not been investigated yet.

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70    Accordingly, this study evaluated the effects of two new types of phosphate fertilizer (UP and APP),

71    one traditional phosphate fertilizer (MKP) on the clogging behavior of four flat emitters at three

72    fertilization concentrations level (0, 0.15 and 0.3 g/L). Hence, the objects of this study were threefold: 1)

73    determine the effectiveness of two new types of phosphate fertilizers (UP and APP) to alleviate the emitter

74    clogging with HSW; 2) clarify the mechanisms emitters clogging using phosphate fertilizer; and 3) identify

75    suitable application mode (i.e. the type and concentration of P fertilizer) of HSW phosphate drip fertigation

76    systems.

## 2. Materials and methods

### 2.1. Experimental setup

79    The experiment was conducted at Tongzhou Experiment Station of China Agricultural University (116°

80    41'2. 31″ E,39° 42'6. 93″ N) in 2018. The saline water source was a well located at the station (Table

81    A1). The drip irrigation with HSW testing system shown in Fig. 1 was divided in four layers, each layer

82    was equipped with one type of emitter and each emitter type facilitated with eight replicates. Four types of

83    flat non-pressure compensating emitters were subjected for the experiment, the detailed physical

84    parameters are as listed in Table A2. The length of each drip lateral was 18 m, the emitters on it with a

85    consistent spacing of 0.33 m in between, thus altogether 55 emitters from inlet to outlet were used. The

86    system operating pressure during the operation was kept as 0.1 MPa, and the amount of water and fertilizer

87    was monitored with a water meter. In this experiment, the phosphate fertilizers were: MKP, UP and APP;

88    fertilizer concentrations were: 0 g/L, 0.15 g/L and 0.30 g/L. Overall, there were 7 treatments, i.e.,

89    MKP_0.30, MKP_0.15, UP_0.30, UP_0.15, APP_0.30, APP_0.15 and CK (non-fertilization). The system

90    was operated once a day, representing the actual field irrigation or fertilization conditions. Fertilization was

91   carried out every other day at the center of the whole operation time. The quantity of applied fertilizer and

92   irrigation water for each drip irrigation lateral was 33.75 g/day and 0.45 m$^3$/day. The fertilization/irrigation

93   plans (Fig. A1) included low concentration/long term (LCLT), high concentration/short term (HCST) and

94   CK treatments.

95                                 #Fig. 1 approximately here#

96   **2.2. Testing method of emitter discharge evaluation**

97   The weighing method (Zhou et al., 2019) was adopted to calculate the emitters' discharge. The

98   multi-layer flow-measuring trolley, which had a 4-layer structure, was used to measure the flow of emitters

99   by placing small buckets (internal diameter of 0.15 m) underneath each tested emitter. A stopwatch (with

100  an accuracy of 0.01 s) was used to record measuring time of 5 min. Finally, the weight measured by a

101  high-precision electronic balance (with an accuracy of $10^{-3}$ g) together with measuring time allowed

102  calculating the flow rate of each emitter.

103  **2.3. Evaluation parameters of emitter performance**

104  Average discharge variation rate (*Dra,* Li et al., 2019b) was computed to determine the clogging

105  degree of emitters and Christiansen uniformity (*CU,* Christiansen., 1942) was used to quantify the

106  irrigation uniformity of several emitters along a lateral (Section 1.5 of the supplementary materials).

107  **2.4. Extraction and evaluation of clogging substances in emitters**

108  During the whole experiment, each treatment was sampled 8 times. At each sampling event, drip

109  lateral was destructively fetched, hereafter 5 samples out of the 15 emitters in head, middle and tail were

110  selected. The samples were placed in centrifuge tubes and stored in refrigerators at 4℃.

111      Dry weight (*DW*) of substances inside emitters determined the total clogging. Fifteen emitter samples

112      were placed in zip lock bags, added 20 mL of deionized water to remove the clogging substances for 60

113      min using the ultrasonic cleaner (manufacture: Chaowei; type: GVS-10 L). The mixed solid phase of 15

114      emitter samples acquired at the bottom was dried to a constant weight and then weighed using a

115      high-precision electronic balance.

116      The dried solid phase was ground and X-ray diffractometer (manufacture: Bruker, Germany; type:

117      D8-Advance) was used to get the polycrystalline diffraction patterns. Basic test conditions for the scanning

118      process were: voltage 40 kV, current 40 mA, Cu target, wavelength $\lambda=1.5406$ Å. Then, the patterns were

119      analyzed by the Topas software (Bruker Corp) to obtain the mineral contents and components of the

120      clogging substances. The polycrystalline diffraction patterns were as Fig. A2 (E1 as a sample).

121      In fertilization treatments, amorphous components were found. The component of amorphous

122      components was tested by Fourier transform infrared spectrometer (FTIR) and scanning electron

123      microscope with an energy dispersive spectrometer (SEM-EDS). The component of amorphous

124      components was phosphate.

125      **2.5. Statistical analysis**

126      The Pearson analysis was applied to study the correlations among the *Dra*, *CU* and mineral contents.

127      Paired t-test was used to analyze the differences between different treatments. During these processes, the

128      significance thresholds of the independent variables were determined at $p < 0.05$ and $p < 0.01$. The

129      regressions and statistical analyses were carried out using SPSS (ver. 20.0, IBM Analytics).

130      # 3. Results

131      **3.1. Effect of phosphorous fertigation on emitter performance (*Dra* and *CU*)**

132 The system *Dra* (Fig. A3) and *CU* (Fig. A4) indicated that emitters' performance across all treatments

133 decreased over time. The performances showed a slow decline during the early stage (0 - 16 d) and a rapid

134 decline during the later stages (16 - 64 d). Moreover, emitters' performance varied greatly among different

135 treatments. The correlations of *Dra* and *CU* between different treatments are shown in Fig. 2. Compared to

136 CK, UP and APP significantly ($p < 0.05$, Table A3) alleviated the emitter clogging. The *Dra* and *CU* of UP

137 treatments were increased by 25.0% - 45.0% and 26.2% - 44.5% and APP treatments increased by 46.3% -

138 73.6% and 46.1% - 74.6%, respectively. Conversely, *Dra* and *CU* of MKP decreased by 14.3% - 34.7%

139 and 10.8% - 38.6%, respectively, meaning that emitter clogging was worsened. Fertigation concentrations

140 also exhibited a significant impact on the emitter clogging (Table A3). APP significantly alleviated the

141 clogging of the emitter under the high-concentration (0.30 g/L) and short-term (HCST) fertilization mode,

142 which increased *Dra* and *CU* by 50.9% and 52.7% than under low-concentration (0.15 g/L) and long-term

143 (LCLT) fertilization mode. But UP and APP treatments showed opposite results. Both *Dra* and *CU* of UP

144 under HCST fertilization mode were significantly lower than LCLT fertilization mode by 36.3% and

145 33.1%, respectively. For MKP, the reduction ratio of *Dra* and *CU* were 17.9% and 25.1%, respectively.

146          #Fig. 2 approximately here#

147 **3.2. Total dry matter (*DW*) and mineral components of emitter clogging substances**

148 Total *DW* of clogging substances across each treatment increased gradually over time. Moreover, the

149 type and concentration of fertilization directly affected the accumulation of *DW* (Fig. A5). Compared to

150 CK, UP and APP significantly ($p < 0.05$) decreased the *DW* of clogging substances (Table A4) by 21.1% -

151 28.6% and 32.2% - 42.0% (Fig. 3). After the application of MKP, the *DW* increased by 3.0% - 18.7%.

152 Significant effect of fertilization concentrations on *DW* was also observed (Table A4). With the increase of

153  fertilizer concentration, APP significantly alleviated more the *DW* of the emitter under the HCST

154  fertilization mode, which decreased *DW* by 14.7% than LCLT fertilization mode. However, the *DW* of UP

155  and MKP under HCST fertilization mode were significantly higher than LCLT fertilization mode by 9.7%

156  and 15.5%.

157    The mineral components inside emitters are shown in Fig. 4 (taken E1 as an example, at the end of the

158  experiment) and Fig. A6 (all emitters and all periods). The results indicated that mineral components

159  mainly included muscovite $[K(Mg,Al)_{2.04}(Si_{3.34}Al_{0.66})O_{10}(OH)_2]$, clinochlore $[(Mg_5Al)(Si_3Al)O_{10}(OH)_8]$,

160  anorthite $[CaAl_2Si_2O_8]$, monohydrocalcite $[CaCO_3 \cdot H_2O]$, dolomite $[CaMgCO_3]$, aragonite $[CaCO_3\text{-}O]$,

161  calcite $[CaCO_3\text{-}R]$, quartz $[SiO_2]$, and phosphate (the determination process of phosphate is shown in

162  Section 2.5 of the supplementary materials). The nine primary mineral components can be divided into

163  carbonate, phosphate, silicate, and quartz according to the main chemical elements. The mineral

164  component of CK was mainly carbonate, and the mineral components for fertilization treatments were

165  mainly a mixture of carbonate and phosphate. The ratios of the four types minerals across all treatments for

166  emitter E1 (Fig. A9) were: 18.5% - 94.1% (carbonate), 34.6% - 60.1% (phosphate, except CK), 5.0% -

167  15.9% (silicate), and 0.9% - 8.2% (quartz), respectively.

168                         #Fig. 3 approximately here#

169                         #Fig. 4 approximately here#

170  **3.3. Correlation of mineral components among different treatments**

171    The variation of average carbonate, phosphate, silicate, and quartz content (Fig. A10) indicated that

172  all mineral components showed a slow increase during early stage (0 - 16 d) and a rapid increase during

173  the later stages (16 - 64 d). Moreover, mineral content varied greatly across treatments. Compared to CK,

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174　UP, APP and MKP all significantly ($p < 0.05$, Table A5) inhibited the formation of carbonate (Fig. A10, Fig.

175　5a). The carbonate content in three fertilizer treatments (UP, APP and MKP) were decreased 50.4% -

176　59.2%, 66.0% - 81.5%, and 74.1 - 76.2%, respectively. Fertilizer concentration had a significant effect ($p <$

177　0.05, Table A5) on carbonate content. Thus, the carbonate content in MKP_0.30 was significantly lower

178　than MKP_0.15 by 17.7%, UP_0.30 was significantly lower than UP_0.15 by 45.5%, but APP_0.30 was

179　significantly lower than APP_0.15 by 8.8%.

180　　　Significant differences ($p < 0.05$, Table A5) were observed in phosphate contents among different

181　treatments (Fig. 5b). The phosphate contents of UP_0.15 and APP_0.15 were lower than MKP_0.15 by

182　18.0% and 5.9%, and UP_0.30 and APP_0.30 were lower than MKP_0.30 by 36.5% and 43.9%. Fertilizer

183　concentration had an evident effect on phosphate content. The phosphate contents in HCST fertilization

184　mode of UP, APP, and MKP were significantly ($p < 0.05$) increased by 40.0%, 7.5% and 80.7%,

185　respectively.

186　　　Compared to CK, the three P fertilizers significantly ($p < 0.05$, Table A5) promoted the formation of

187　silicate (Fig. 5c). The silicate content of the UP, APP, and MKP increased 46.6% - 102.8%, 21.3% - 75.8%

188　and 56.1% - 234.9% regarding CK, respectively. Fertilizer concentrations also had a significant ($p < 0.05$)

189　effect on silicate content. The silicate content of MKP_0.30 was lower than MKP_0.15 by 53.4%, UP_0.30

190　was lower than UP_0.15 by 31.0%, and APP_0.30 was lower than APP_0.15 by 27.8%.

191　　　The quartz content in different treatments (Fig. 5d) was also significantly different ($p < 0.05$, Table

192　A5). The quartz content of the UP, APP, and MKP increased 218.3% - 462.2%, 146.1% - 396.3% and 448.2%

193　- 657.6%. The quartz content of MKP_0.30 was 38.2% higher than MKP_0.15, UP_0.30 was 76.6% lower

194　than UP_0.15, but APP_0.30 was 50.4% lower than APP_0.15.

195　　　　　　　　　　　　　　　#Fig. 5 approximately here#

**3.4. Correlation between the content of mineral components and *CU***

A linear relationship between *Dra* and *CU* were found (Fig. A11). The results shown in Fig. 6 (a) - (d) indicated that the contents of carbonate, phosphate, silicate and quartz in all treatments were significantly negatively correlated ($p < 0.05$, $R^2 > 0.82$) with their *CU*, and their fitting parameter K (regression line slope coefficient) for carbonate, phosphate, silicate and quartz ranged -5.28 - -0.96, -5.53 - -0.66, -27.01 - -6.89 and -124.34 - -15.19.

#Fig. 6 approximately here#

# 4. Discussion

**4.1. Mechanism of phosphorous fertilizer on emitter clogging**

The results of this experiment (Fig. 2) showed that the application of UP and APP could effectively relieve the emitter clogging in drip irrigation system with HSW. UP alleviated the emitter clogging by reducing the content of carbonate, although the increased content of phosphate, silicate, and quartz in mineral components of emitter clogging substances (Fig. 4). The decrease of carbonate is attributed to the acidic nature of the UP solution, as produced of 6.3 mol $H^+$ for 1 kg of dissolved UP, which promotes the transformation of $CO_3^{2-}$ to $HCO_3^-$ and thus inhibits the formation of carbonate. In addition, the hydrolysis of UP produced a large amount of $PO_4^{3-}$, which competed with $CO_3^{2-}$ for $Ca^{2+}$ and $Mg^{2+}$. Therefore, the production of calcium phosphate precipitate was promoted (Fig. 4), showing a power function growth trend, and the production of carbonate precipitate was further inhibited (Hammes and Verstraete, 2002). Moreover, the application of P reduces the pH (1% UP aqueous solution has pH 1.89) of HSW, resulting in a significant increase in silicate contents (Table A5). Actually, pH decrease promoted the condensation of silanol structure, and impelled the polymerization of silicate gel (Neofotistou and Demadis, 2004), and

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217 thus exacerbated silicate deposition. The increase in quartz was probably due to the negative charge of

218 particulate matter such as quartz in water source. While the addition of UP fertilizer induced a lot of

219 positive charge to compress the electric double layer by electrostatic attraction and became thinner when

220 the cations increased and zeta potential decreased (Zhou et al., 2019), leading to significant increase in the

221 solid particulates' flocculation and sedimentation strength (Liu et al., 2017). Since the experiments were

222 carried out at room temperature and 100 kPa pressure, it is difficult to form $SiO_2$ in the emitter at these

223 conditions. Therefore, the $SiO_2$ found in clogging substances were mainly induced from the water (Zhou et

224 al., 2018). Taking quartz as an independent variable, the accumulation processes of phosphate, carbonate

225 and silicate (as dependent variables) are plotted in Fig. A12. The results showed that there was a high

226 linear relationship between silicate and quartz, which indicated that the variation rate of silicate was

227 relatively stable along with system operation. The stable variation further revealed that silicate and quartz

228 are basically homologous, originated from water, rather than resulted from a chemical reaction during

229 process of system operation. The carbonates and phosphates showed a power-function growth, which

230 indicated that not only came with water source but also resulted from chemical reactions.

231 APP treatment also significantly reduced the emitter clogging. Similar to UP, the content of carbonate

232 in the emitter clogging substances when using APP decreased significantly, while silicate and quartz

233 increased, and phosphate component were appeared at the same time. APP also produced competitive

234 adsorption to form phosphate, which reduced the content of carbonate (Fig. A10). However, APP did not

235 significantly change the pH of fertigation water (Table A5). The reason why the content of carbonate

236 decreases greatly is APP is usually a mixture of different chain lengths (Waerstad and McClellan, 1976).

237 This special structure results in that APP can chelate $Ca^{2+}$ and $Mg^{2+}$ and other metal ions at a certain

238 concentration, will shield $Ca^{2+}$ and $Mg^{2+}$ (Noy and Yoles, 1979; Hagin et al., 2002), thus reducing the

239     probability of carbonate reacting with $Ca^{2+}$ and $Mg^{2+}$, and greatly reducing the formation of carbonate. The

240     formation of phosphate mainly due to the large amount of $PO_4^{3-}$ brought by APP, that inevitably react with

241     calcium ions in water to form insoluble calcium phosphate compounds (Ryan and Saleh., 2000). The

242     formation mechanism of silicate and quartz were similar to UP. Fig. A12 (d) indicated a linear growth trend

243     of both silicate and quartz, while carbonate and phosphate showed a quadratic function growth trend with a

244     gradual slowed-down growth rate, which indicated that APP could inhibit the formation of salt substances.

245        After the application of MKP, emitter clogging drastically increased, while the content of carbonate

246     noticeably reduced. Similar to UP, this was mainly due to the formation of phosphate precipitate by

247     reducing water pH and competitive adsorption of calcium and magnesium ions. Besides phosphate

248     precipitations, MKP increased the silicate and quartz levels in clogging substances (Figs. 4 and 5). During

249     the application of MKP in drip irrigation, large amount of $Ca^{2+}$ and $Mg^{2+}$ in high saline water consumed

250     the $HCO_3^-$ to form a large amount of carbonate, and then the remaining $Ca^{2+}$, $Mg^{2+}$, and $PO_4^{3-}$ produced by

251     the electrolysis of MKP reacted to form phosphate. Dicalcium phosphate was considered to be the main

252     precipitate formed by the reaction of $PO_4^{3-}$ and $Ca^{2+}$ in irrigation water (Chow and Eanes, 2001). Then,

253     dicalcium phosphate hydrolyzed as hydroxyapatite or dicalcium phosphate quickly transformed to

254     octacalcium phosphate and further transformed to hydroxyapatite (Tung and Skrtic, 2001). On the other

255     hand, $CaCO_3$ absorbed $PO_4^{3-}$ and formed insoluble compounds (Zhou et al., 2018). Therefore, under MKP

256     application in high salinity groundwater, the formation of phosphate in water mainly depended on the

257     adhesion of attachment of $PO_4^{3-}$ on $CaCO_3$ surface and ion reaction (Bertrand et al., 2006). At the same

258     time, the deposition of phosphate and carbonate greatly increased the surface area of clogging substances

259     on the inner surface of the emitter, which further promoted the deposition of clogging substances on the

260     surface of flow channel (Zhou et al., 2019). The two effects together resulted in the exponential growth

261    trend of carbonate and phosphate, while the content of silicate and quartz increased linearly (Fig. A12 b),

262    which together aggravated the clogging of emitters.

263        It was found that the concentration of phosphorus fertilizer would affect the clogging rate of the

264    emitter under the same amount of fertilizer. UP and APP could effectively alleviate the clogging of emitter

265    under the mode of LCLT and HCST, respectively. For UP treatment, the clogging degree of HCLT

266    fertilization treatment was significantly higher than LCLT fertilization treatment. This was mainly because

267    the phosphate and quartz precipitation were significantly increased under HCLT fertilization mode. The

268    increase of phosphate may be related to time factors, the longer the reaction time was, the more phosphate

269    formed (Bremner and Douglas, 1971). The higher the concentration of UP, the more noticeable the

270    flocculation and sedimentation of quartz, and the higher the content of quartz (Liu et al., 2017). For APP

271    treatment, the higher the fertilizer concentration, the lower the risk of emitter clogging. With the increase

272    of APP concentration, the chelating effect gradually increased, and the effect of reducing carbonate

273    precipitation gradually appeared. APP also reacted with $Ca^{2+}$ and $Mg^{2+}$ at a certain concentration to form a

274    colloidal suspension clogging the emitter, which is related to the content of $Ca^{2+}$ in the water source and

275    the concentration of ammonium polyphosphate (Noy and Yoles, 1979). Hagin and Tucker (1982) indicated

276    that injection of small quantities of ammonium polyphosphate solution into water high in dissolved

277    calcium resulted in precipitation of calcium ammonium pyrophosphate, while injection of large quantities

278    of polyphosphate caused the precipitate to disappear due to the sequestering ability of the polyphosphate.

279    Duis and Burman (1969) found that for ammonium polyphosphate (11-37-0) in a water containing up to

280    100 mg/L Ca no precipitate will form regardless of the ammonium polyphosphate added to supply a high or

281    low concentration in the irrigation water, whereas in a water containing 200 mg/L Ca the polyphosphate in

282    a 1:300 dilution in water will form precipitates, but a 1:200 dilution will prevent their formation.

**4.2. Selection of phosphate fertilizer application mode**

Although demonstrated the effect of phosphorus fertilizer application on the clogging of drip irrigation emitter, the effects of soil texture, fertilizer price and availability need to be considered in the practical application of phosphate fertilizer at farm level (Hagin and Tucker, 1982; Goyal and Mansour., 2015b).

Firstly, in addition to alleviating emitter clogging, low concentration UP can effectively reduce soil pH which is suitable to apply UP in alkaline soil (Mikkelsen and Jarrell, 1987). Secondly, UP can promote the conversion of phosphate to $H_2PO_4^-$ form, thus enhance phosphorous availability and use efficiency (Kafkafi and Tarchitzky, 2011). Thirdly, the price of UP is relatively low, the purchasing prices of UP, PPM and APP mainly ranged \$650 - 840 /t, \$870 - 1090 /t and \$945 - 1310 /t (China fertilizer website, 2019), respectively. Finally, UP can promote the early flowering and fruiting of crops (Becker et al., 2004), as early market entry is of great benefit to raising the price of agricultural products. However, due to excessive fertilization and other reasons, the soil in many areas presents the risk of continuous acidification (Tian and Niu, 2015).

Continuous application of UP may further reduce soil pH (Mikkelsen, 1989), thereby threatening the crop growth. Therefore, for acidic and neutral soils, APP might be more suitable through drip irrigation systems with HSW (Kafkafi et al., 2011). The price of fertilizer is also one of the main factors to be considered since the price of APP is comparatively higher. Improper application of APP may lead to the situation of increasing production while not increasing income (Noy and Yoles, 1979). In the case of low-volume drip irrigation of phosphate fertilizer, because of its lower cost and higher fertilizer efficiency, APP fertilizer is more affordable than top-grade packaged monoammonium phosphate (Ottman et al., 2006). However, at present, there are still few agricultural ammonium polyphosphate manufacturers on

305 several agricultural markets such as China, the price is also relatively expensive and the product quality is

306 not very stable (Yin et al., 2018). Therefore, it is urgent to optimize the APP production process and reduce

307 the price.

308     In general, low concentration UP treatment and high concentration APP treatment significantly

309 reduced the risk of emitter clogging. The mechanism of these two new phosphate fertilizers was clarified

310 for reducing emitter clogging using HSW, and put forward the suitable fertilization mode of UP and APP.

311 However, the effects of local soil type, fertilizer cost and availability should also be considered.

312 Meanwhile, there are two aspects worth exploration in the future: (1) The effects of different phosphorus

313 fertilizer types on-site soil conditions and crop growth under drip irrigation with HSW, (2) Both UP and

314 APP slow down the emitter clogging by inhibiting the formation of carbonate and increase the risk of

315 silicate. Therefore, different results may be produced for water sources with different compositions, e.g.,

316 more silicates, less calcium and magnesium, etc.

## 317 5. Conclusions

318     (1) The application of UP and APP can effectively alleviate the emitter clogging in drip fertigation

319 systems with HSW. Compared with non-fertilization, *Dra* was increased by 25.0% - 45.0% and 46.3% -

320 73.6%, *CU* increased by 26.2% - 44.5% and 46.1% - 74.6%, and *DW* decreased by 21.1% - 28.6% and

321 32.2% - 42.0% with UP and APP, respectively. In contrast, MKP accelerated emitter clogging issues.

322     (2) Phosphate fertilizer significantly changed the content of different minerals inside emitters.

323 Compared with non-fertilization, the carbonate content of MKP, UP and APP decreased by 50.4% - 59.2%,

324 66.0% - 81.5% and 74.1 - 76.2%, silicate increased by 56.1% - 234.9%, 46.6% - 102.8% and 21.3% -

325  75.8%, quartz content increased by 448.2% - 657.6%, 218.3% - 462.2% and 146.1% - 396.3%,

326  respectively. Phosphate precipitation was found in all the clogging materials treated by fertilization.

327      (3) Different fertilization concentration significantly affected emitter clogging. Compared with the

328  UP_0.15, *Dra* and *CU* of UP_0.30 decreased by 36.3% and 33.1%, *DW* increased by 9.7%. The *Dra* and

329  *CU* of APP_0.30 increased by 50.9% and 52.7% and *DW* decreased by 14.7%. It is recommended that UP

330  should choose low concentration and long term (LCLT) and APP should choose high concentration and

331  short term (HCST) as the fertilization mode of high saline drip irrigation system.

## Acknowledgements

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454

455

## Captions for Figures and Tables in the Paper

457     Fig. 1. Layout of HSW drip irrigation experimental system.

458     Fig. 2. Correlation of emitter performance parameters between CK and fertilization treatments.

459     Fig. 3. Correlation of $DW$ between CK and fertilization treatments.

460     Fig. 4. Mineral components in each treatment obtained at the end of the experiment (E1).

461     Fig. 5. Correlation of mineral constituent treatments.

462     Fig. 6. Correlation between the mineral composition and $CU$ for the different emitters.

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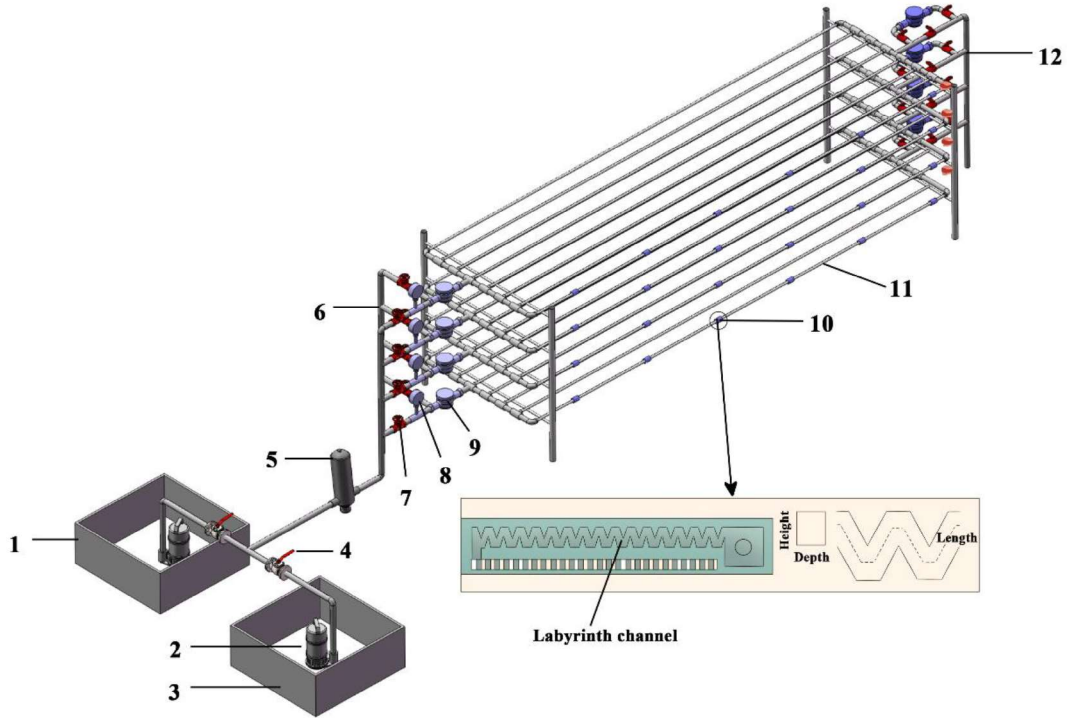


466

467    1 - Water tank, 2 - Water pump, 3 - Fertigation tank, 4 - Butterfly valve, 5 - Disc filter, 6 - Pipes, 7 - Sub

468    valve, 8 - Pressure gauge, 9 - Flowmeter, 10 - Drip emitter, 11 - Drip lateral, 12 - Flushing devices.
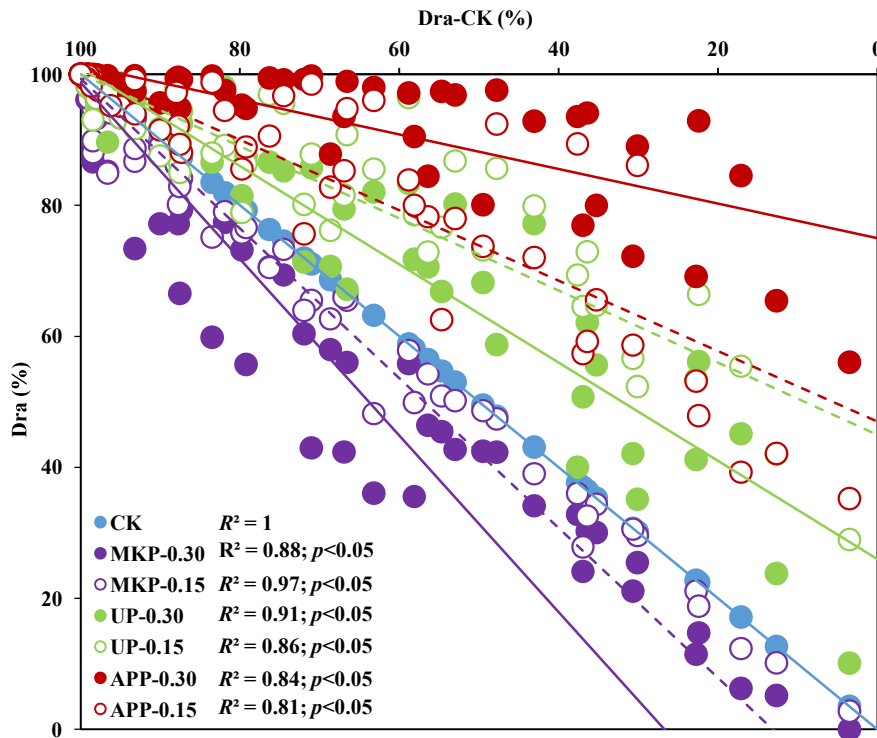
**Fig. 1.** Layout of HSW drip irrigation experimental system.



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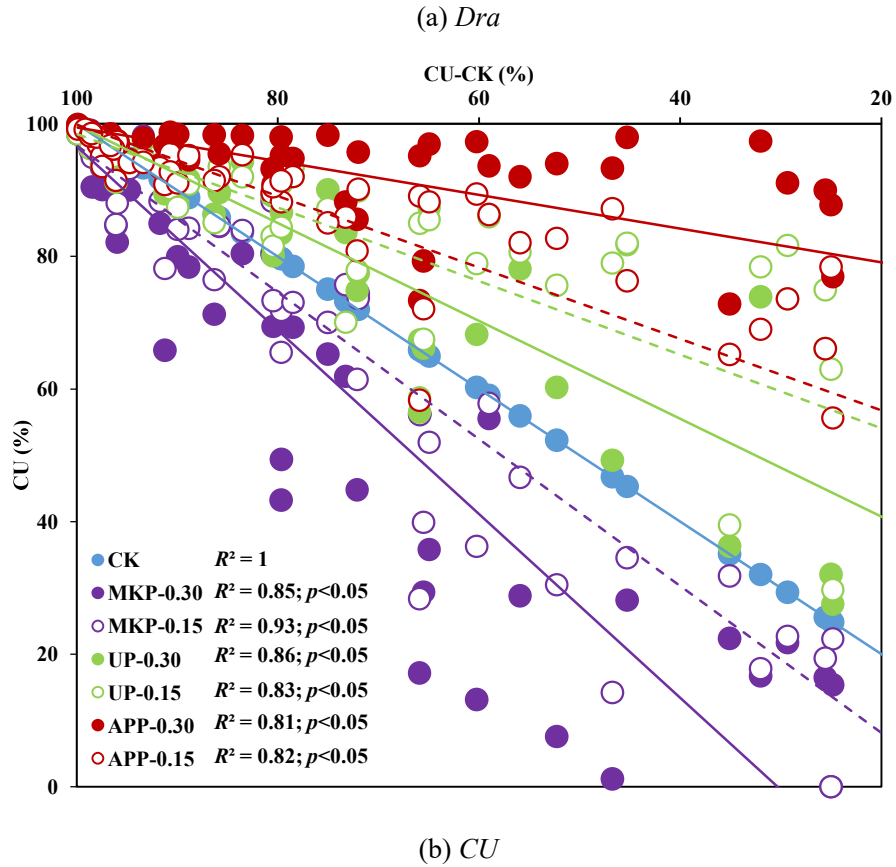
471

(a) *Dra*



472

473

(b) *CU*

**Fig. 2.** Correlation of emitter performance parameters between CK and fertilization treatments.

Note: The x - axis is the *Dra* or *CU* of CK, and the y - axis is the *Dra* or *CU* across different treatments. *p* < 0.05 indicate the treatments are significantly different. The detailed variations for *Dra* and *CU* are shown in Fig. A3 and Fig. A4.
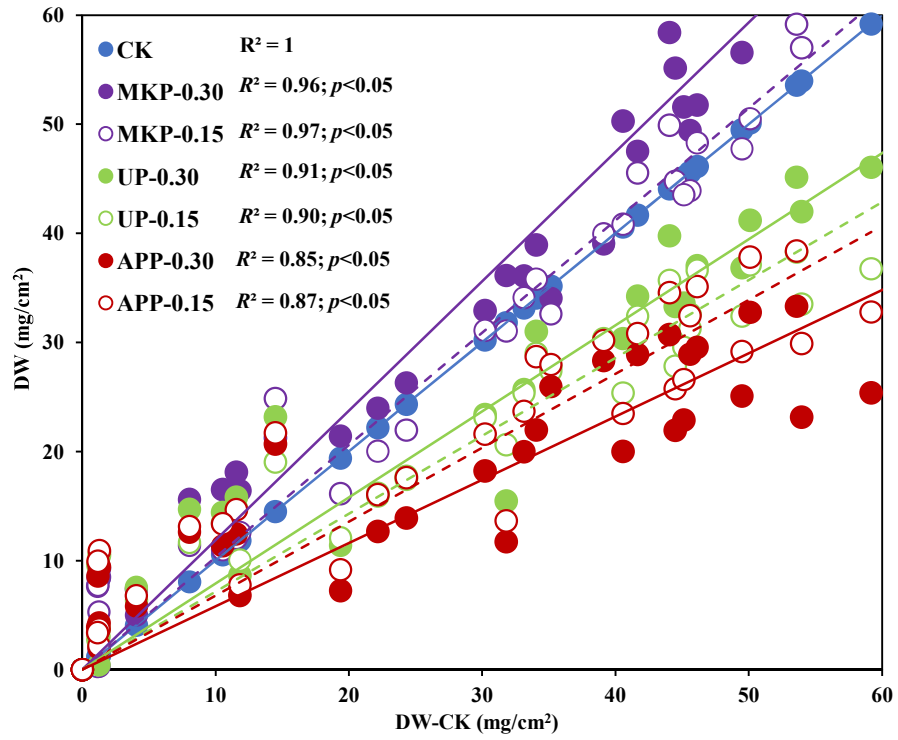
**Fig. 3.** Correlation of *DW* between CK and fertilization treatments.

Note: The detailed variation of *DW* is shown in Fig. A5.
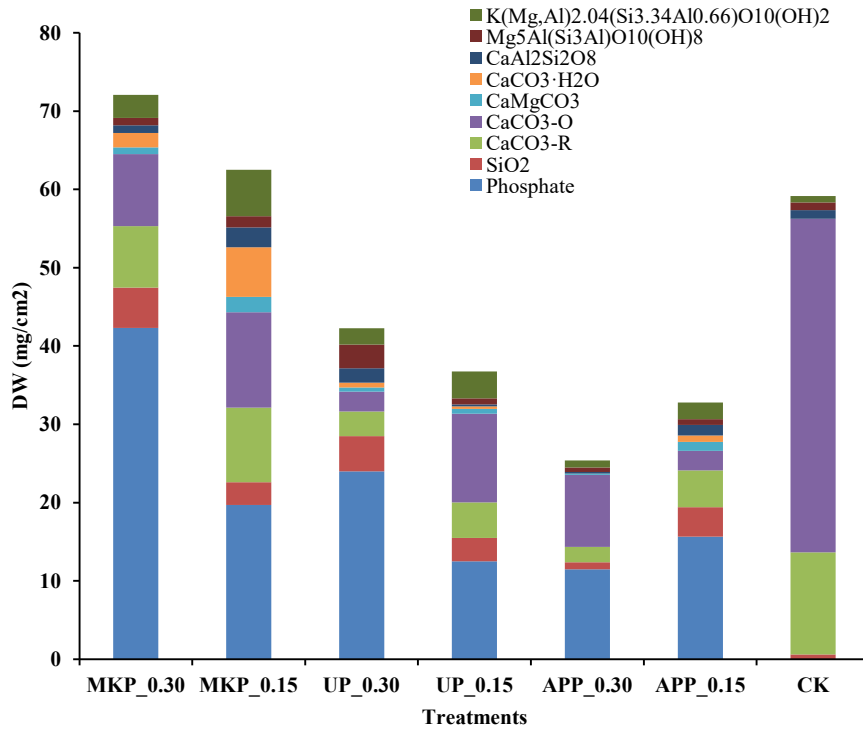
**Fig. 4.** Mineral components in each treatment obtained at the end of the experiment (E1)

Note: As the mineral components were similar across different types of emitters and different periods, only results of FE1 at the end of the periods are shown as an example. Other emitters and periods are shown in Fig. A6. The proportions of each mineral component are shown in Fig. A9.

488

489                                    (a)  Carbonate



490
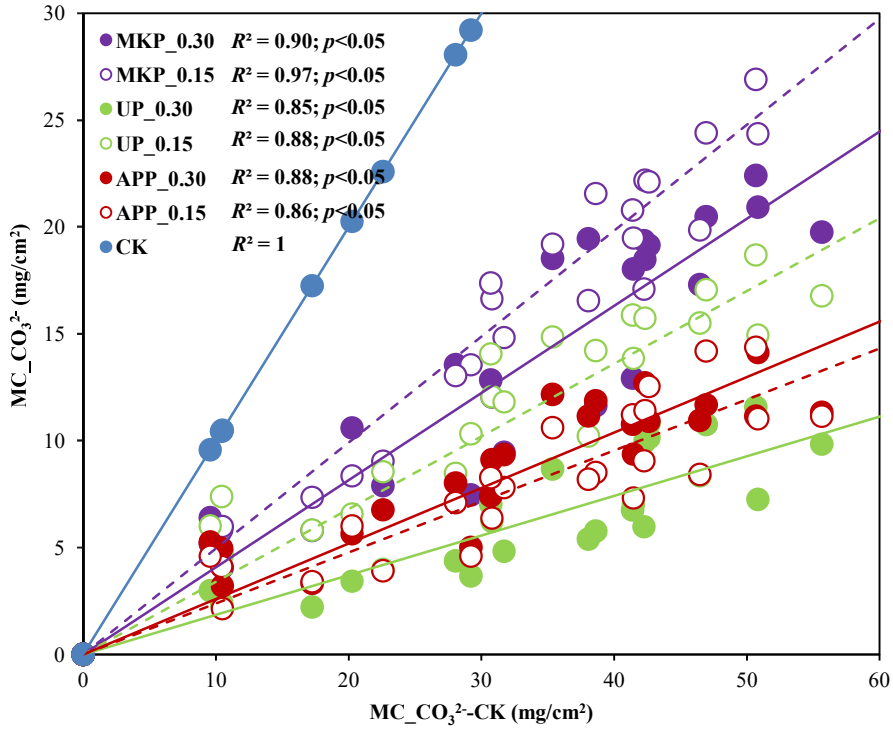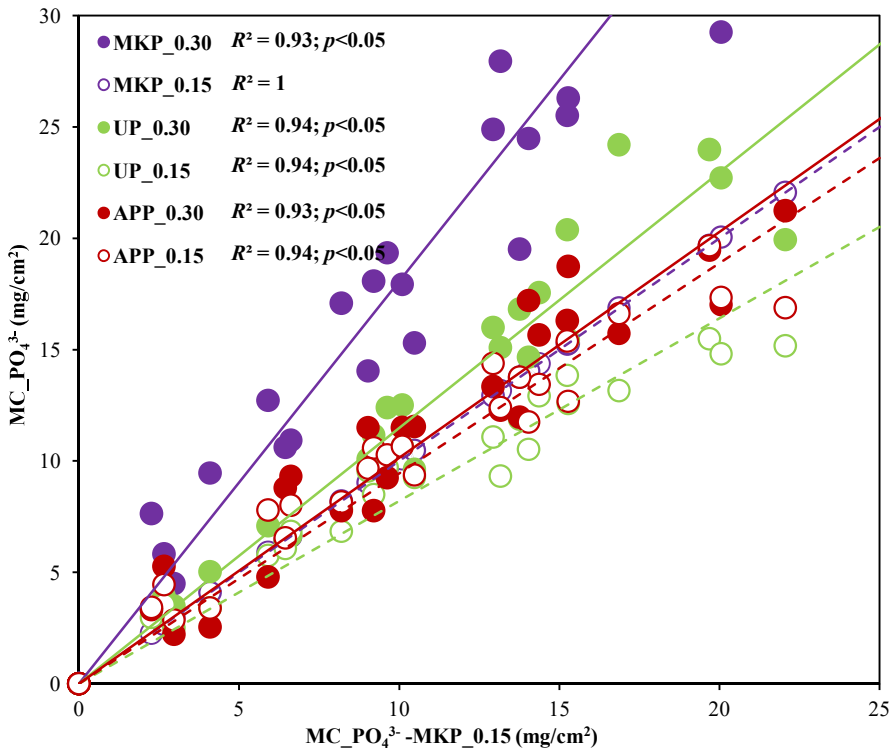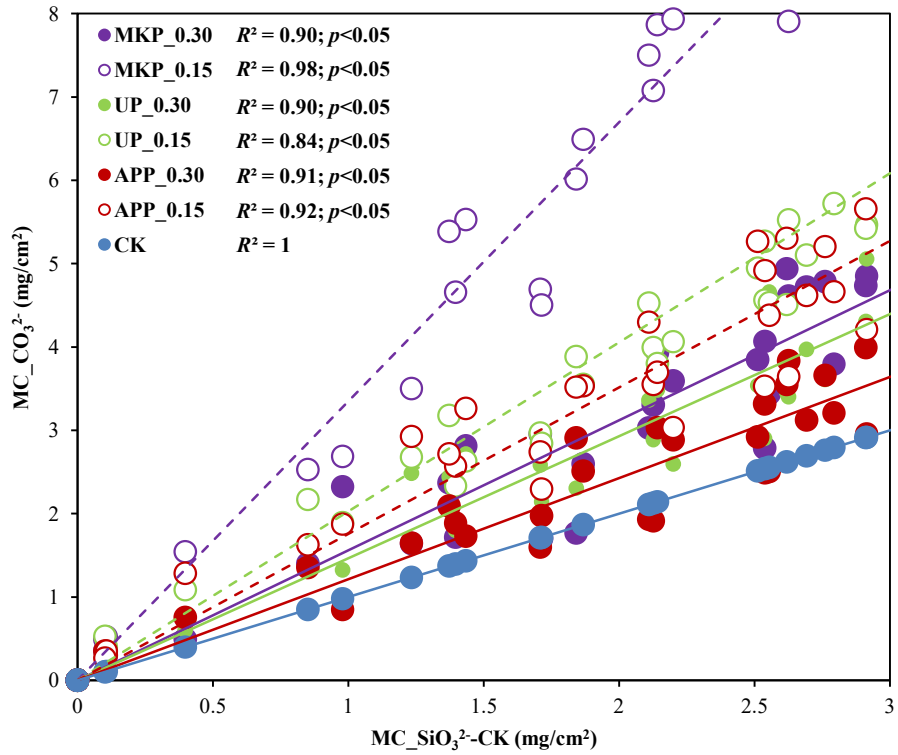
491                                    (b)  Phosphate

492

493                                                  (c) Silicate



494
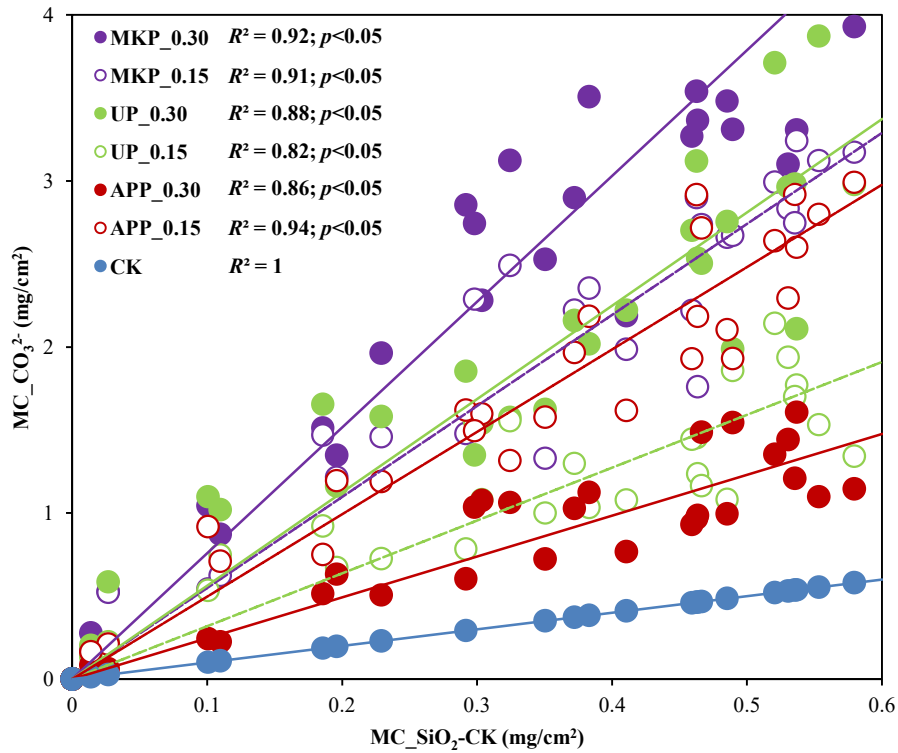
495                                                  (d) Quartz

496                                 **Fig. 5.** Correlation of mineral constituent treatments.

497      Note: The detailed variations of minerals are shown in Fig. A10.
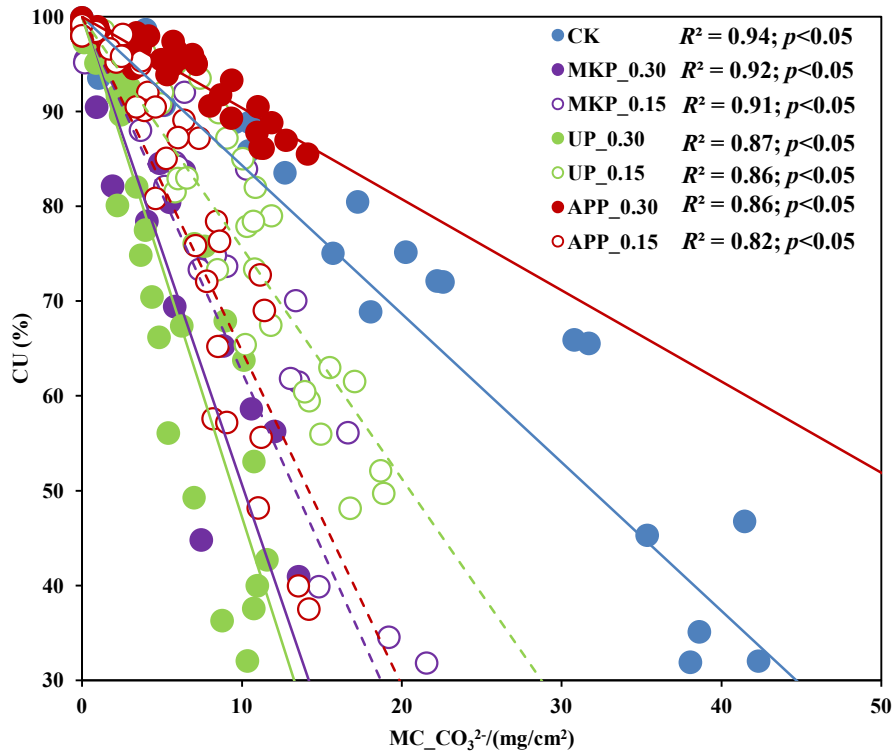
498

499

(a) Carbonate
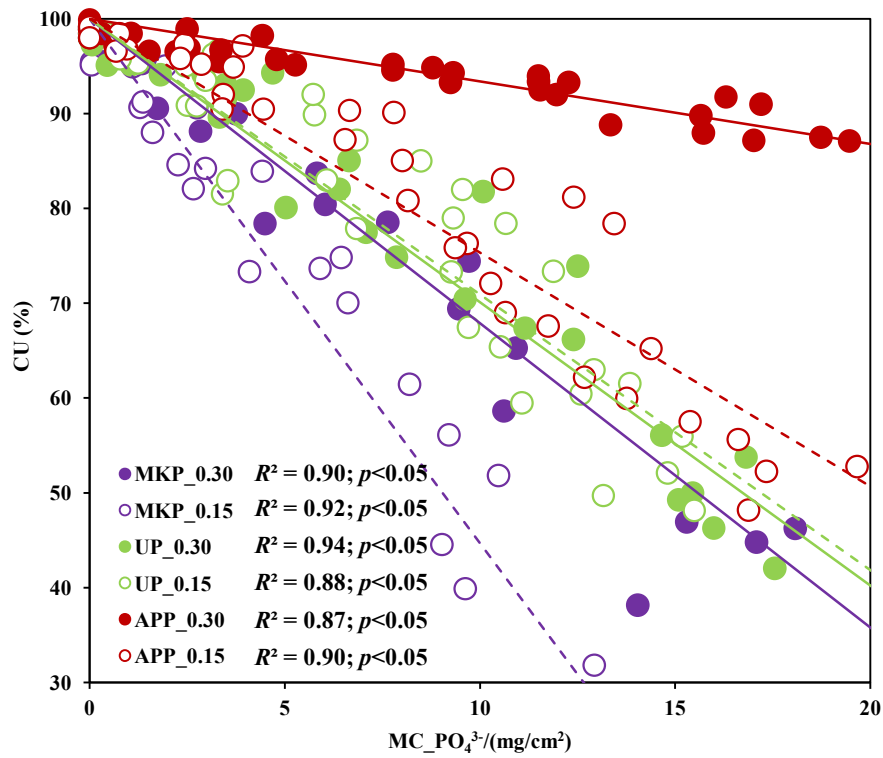


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501

(b) Phosphate
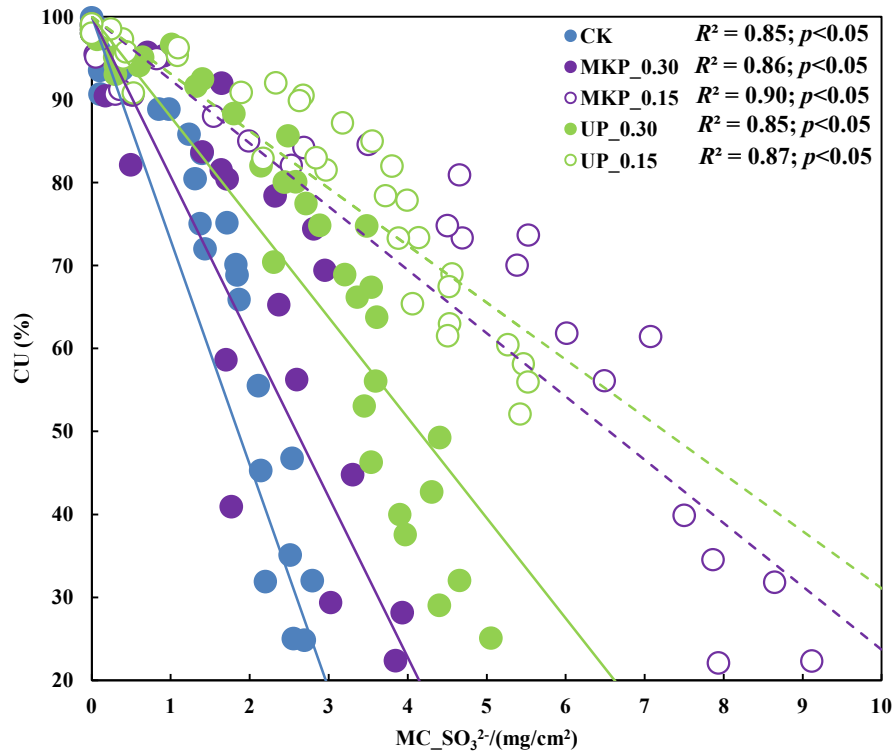
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503

(c) Silicate
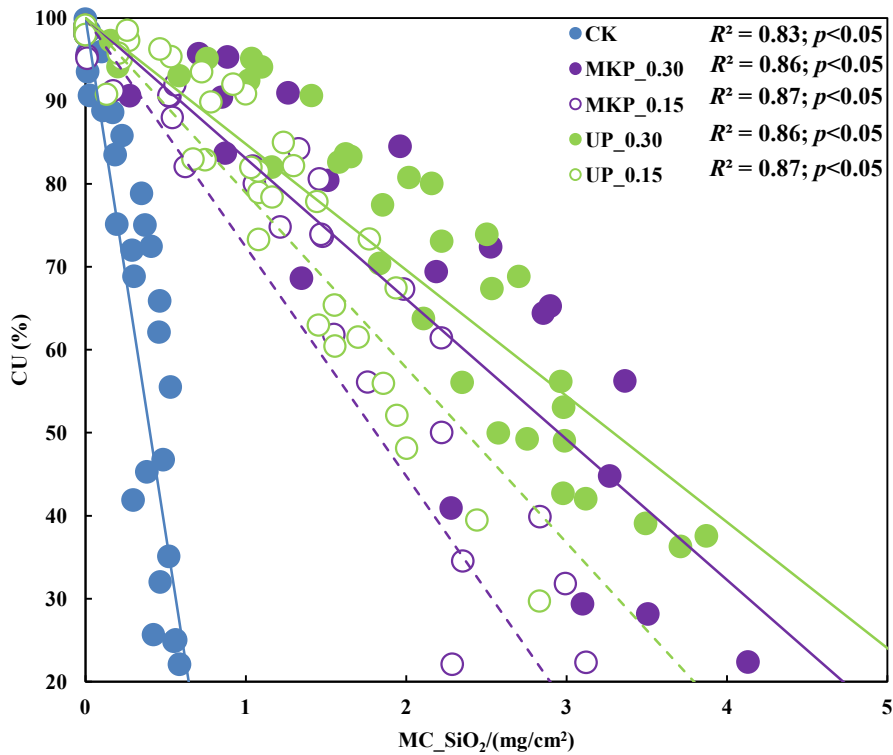


504

505

(d) Quartz

506 **Fig. 6.** Correlation between the mineral composition and *CU* for the different emitters.

507

# SUPPLEMENTARY MATERIALS for

# Using Phosphate Fertilizer to Reduce Emitter Clogging of Drip Fertigation Systems with High Salinity Water

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# 1. Supplementary information in *Experimental section*

## 1.1. Preparation of phosphate fertilizer

The UP was produced by continuous thermal manufacturing process. The main reaction conditions were as follows: the concentration of phosphoric acid was 80% -85 %, the molar ratio of phosphoric acid to urea was 1:1.00 - 1.05, the reaction temperature was 75 - 85 ℃, and the reaction time was 30 min. Its product index was $P_2O_5$ 45.12%, N 17.24%. Based on the preparation of UP, APP was produced by urea phosphate-phosphorus pentoxide polymerization. The ratio of UP to phosphorus pentoxide was 4.44:1, the reaction temperature was 218.93 ℃, and the reaction time was 70.82 min. Under these conditions, the polymerization degree of APP was 23.64, and the solubility was 2.24 g/100g $H_2O$. MKP was produced by neutralization method. Add 30% potassium carbonate solution to the neutralizer, then add 50% phosphoric acid solution and stir. The reaction temperature was 75 - 85 ℃, and pH was ranged 4 - 5. The neutralized product was filtered, concentrated, cooled, crystallized, centrifuged and dried to obtain the finished product. The crystallization mother liquor was returned to the concentration section for reuse. UP, APP, and MKP were produced at Shandong Kingenta Eco-engineering Co., Ltd.

## 1.2. Ranges of water quality parameters

Table A1. Ranges of water quality parameters.

| pH | $Ca^{2+}$ | $Mg^{2+}$ | $Fe^{3+}$ | $Mn^{2+}$ | TH | TDS |
|---|---|---|---|---|---|---|
| | mg/L | mg/L | mg/L | mg/L | mg/L | mg/L |
| 7.50 - 7.55 | 77 - 85 | 78 - 86 | 0.051 - 0.054 | 0.114 - 0.132 | 518 - 535 | 1016 - 1058 |

Note: Water quality parameters were tested by Pony Testing International Group Co., Ltd., China. pH was measured by pH meter. $Ca^{2+}$, $Mg^{2+}$, $Fe^{3+}$ and $Mn^{2+}$ were tested by inductively coupled plasma optical emission spectrometry. Total hardness was tested by burettes. Total dissolved solids were tested by electric blast drying oven and analytical balance.

## 1.3. Geometrical characteristic of emitters used in the experiment

Table A2. Geometrical characteristic of emitters used in the experiment.

| Emitter | Rated discharge(L/h) | Flow path dimensions (mm) | | | Discharge coefficient | Flow index |
|---|---|---|---|---|---|---|
| | | Length | Width | Depth | | |
| E1 | 0.95 | 85.0 | 0.55 | 0.51 | 3.10 | 0.51 |
| E2 | 1.40 | 47.0 | 0.56 | 0.55 | 4.90 | 0.51 |
| E3 | 1.60 | 19.0 | 0.55 | 0.49 | 5.20 | 0.51 |
| E4 | 2.00 | 24.5 | 0.61 | 0.60 | 6.50 | 0.61 |

Note: The flow path dimensions were obtained by an electron microscope after peeling the emitters, and the rated discharge, discharge coefficient, flow index were tested according to China standard (GB/T17188 - 1997).

## 1.4. Drip fertigation/irrigation schedules
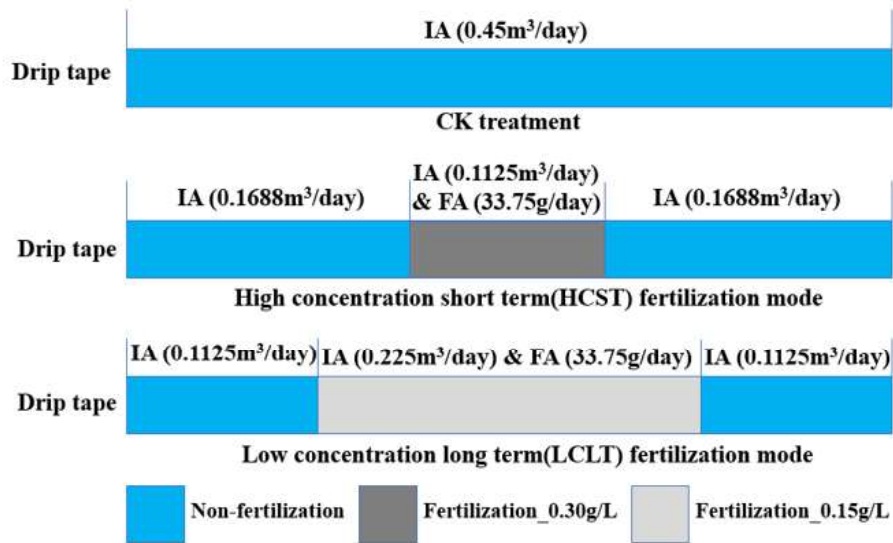


Fig. A1. Drip irrigation system composition Fertigation and/or irrigation schedules with different fertilizer concentrations.

Note: IA, irrigation amount; FA, fertilization amount. Fertigation_0.30 g/L and Fertigation_0.15 g/L represent that the concentrations of fertilizer were 0.30 g/L and 0.15 g/L, respectively. The IA and FA were the same in two fertigation modes (HCST and LCLT), and the amount of irrigation and fertilization per drip irrigation lateral (18 m) was 0.45 $m^3$/d and 33.75 g/d respectively. The two fertigation modes were adopting the "(1) Non-fertilization (Only irrigation); (2) Fertigation; and (3) Non-fertilization" operating schedules, and the fertilization was applied at the center of the whole operation time. Besides, the fertigation treatments were not applied every day, the system fertigation ran every other day.

## 1.5. Evaluation parameters for emitter performance (*Dra* and *CU*)

The theoretical maximum and minimum values of *Dra* were 100% (initial value, non-clogging) and 0% (complete-clogging), respectively. *Dra* was calculated as follows.

$$Dra = \frac{\sum_{i=1}^{n} \frac{q_i^t}{q_i^0}}{n} \times 100\% \tag{1}$$

Where $q_i^0$ is initial flow rate for emitter i, in L/h; $q_i^t$ is flow rate for emitter i at *t* days, in L/h; and *n* is the total number of emitter installed along the lateral.

*CU* values were less than 100% (100% indicates all the emitters along a lateral has same flow rate). *CU* was calculated follows.

$$CU = 100 \left(1 - \frac{\sum_{i=1}^{n} |q_i^t - \bar{q}^t|}{n\bar{q}^t}\right) \tag{2}$$

Where $\bar{q}^t$ is average flow of each emitter along the lateral at t time, in L/h; and other parameters are defined in Eq. (1).

## 1.6. Diffraction pattern of crystal
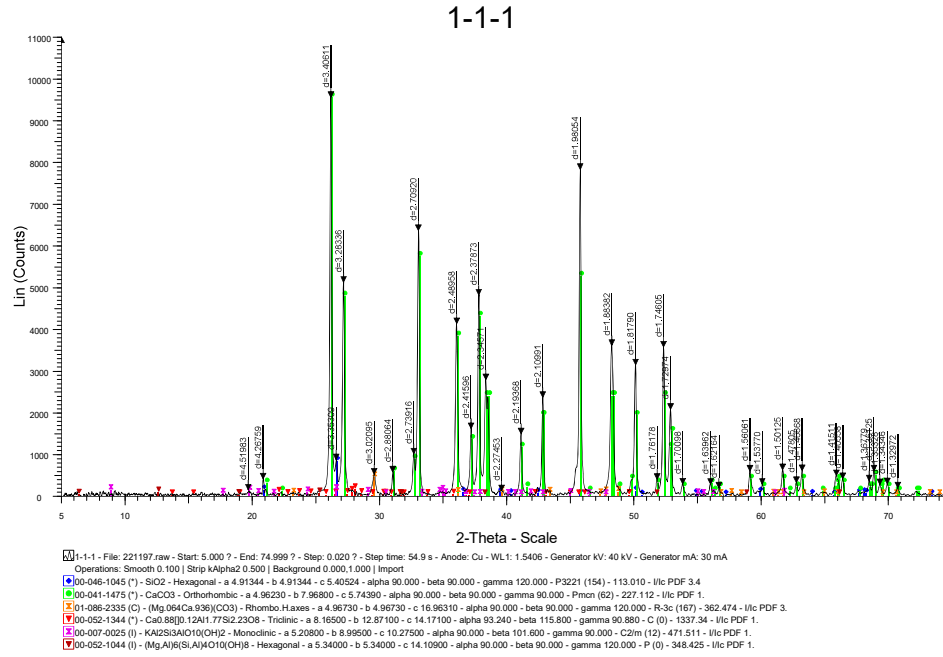


Fig. A2. Polycrystalline diffraction patterns of the emitter clogging substances (E1 as an example).

# 2. Supplementary information in *Results section*

## 2.1. Dynamic variation of *Dra*
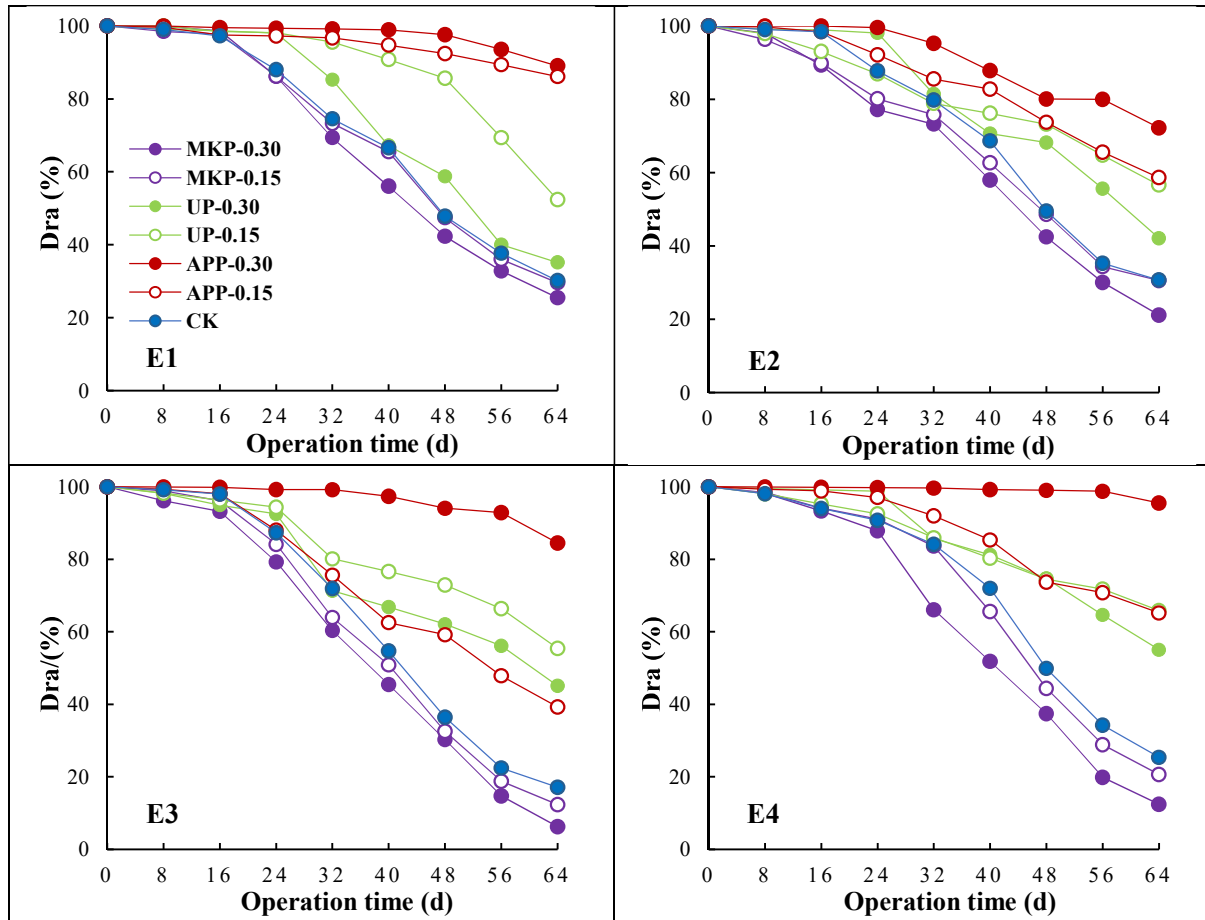


Fig. A3. Dynamic variation of *Dra* for the different emitters (E1-E4) and fertilization treatments.

## 2.2. Dynamic variation of *CU*
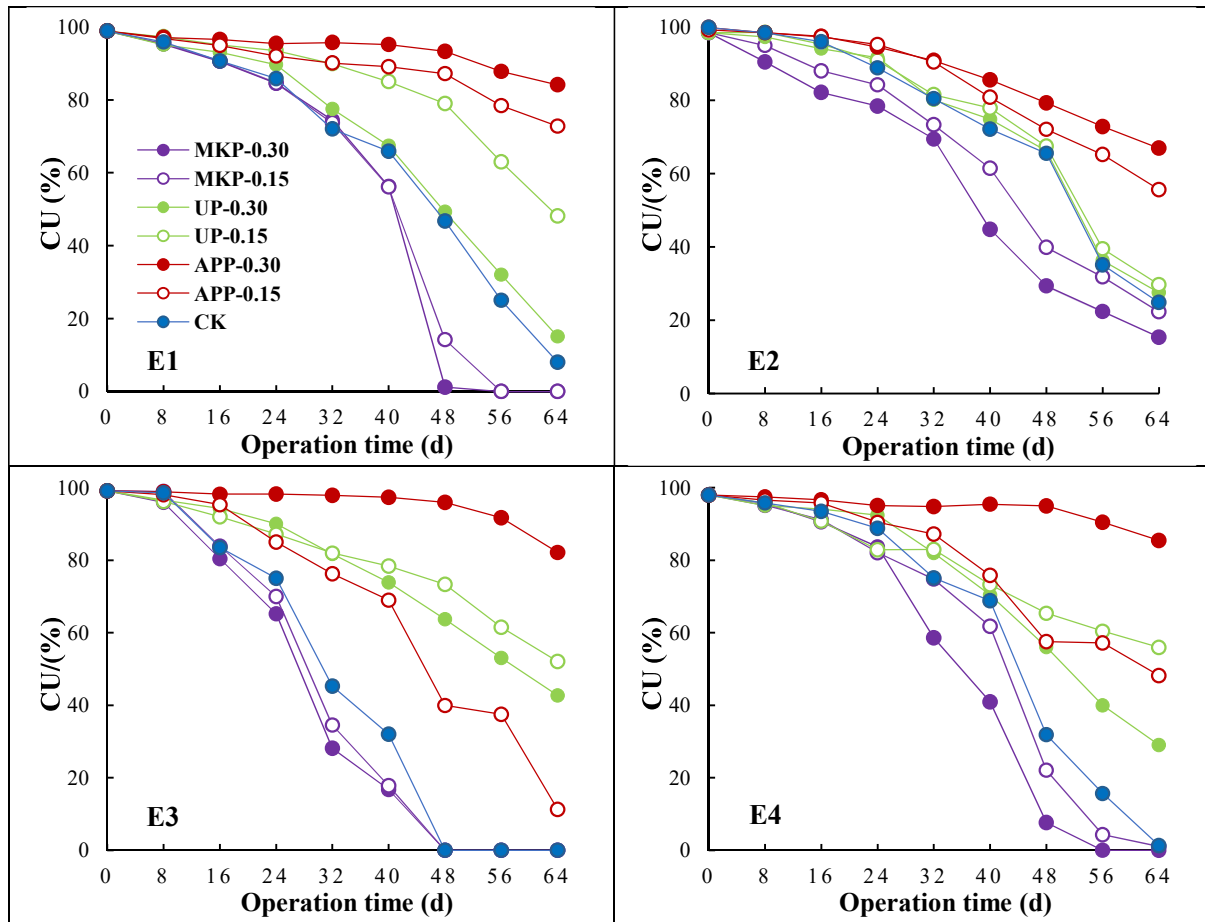


Fig. A4. Dynamic variation of *CU* for the different emitters (E1-E4) and fertilization treatments.

Table A3. Paired samples t-test of emitter clogging parameters in different treatments.

| Treatments | CK | MKP_0.15 | UP_0.15 | APP_0.15 | MKP_0.30 | UP_0.30 | APP_0.30 |
|---|---|---|---|---|---|---|---|
| CK | — | ** | ** | ** | ** | ** | ** |
| MKP_0.15 | ** | — | ** | ** | ** | ** | ** |
| UP_0.15 | ** | ** | — | NS | ** | ** | ** |
| APP_0.15 | ** | ** | NS | — | ** | * | ** |
| MKP_0.30 | ** | ** | ** | ** | — | ** | ** |
| UP_0.30 | ** | ** | ** | * | ** | — | ** |
| APP_0.30 | ** | ** | ** | ** | ** | ** | — |

Note: asterisks indicate that differences between treatments are significant at $p < 0.05$ (*) or $p < 0.01$ (**), and NS means not significant at $p < 0.05$. The upper right is *Dra*, the lower left is *CU*.

## 2.3. Dynamic variation of dry matter weight
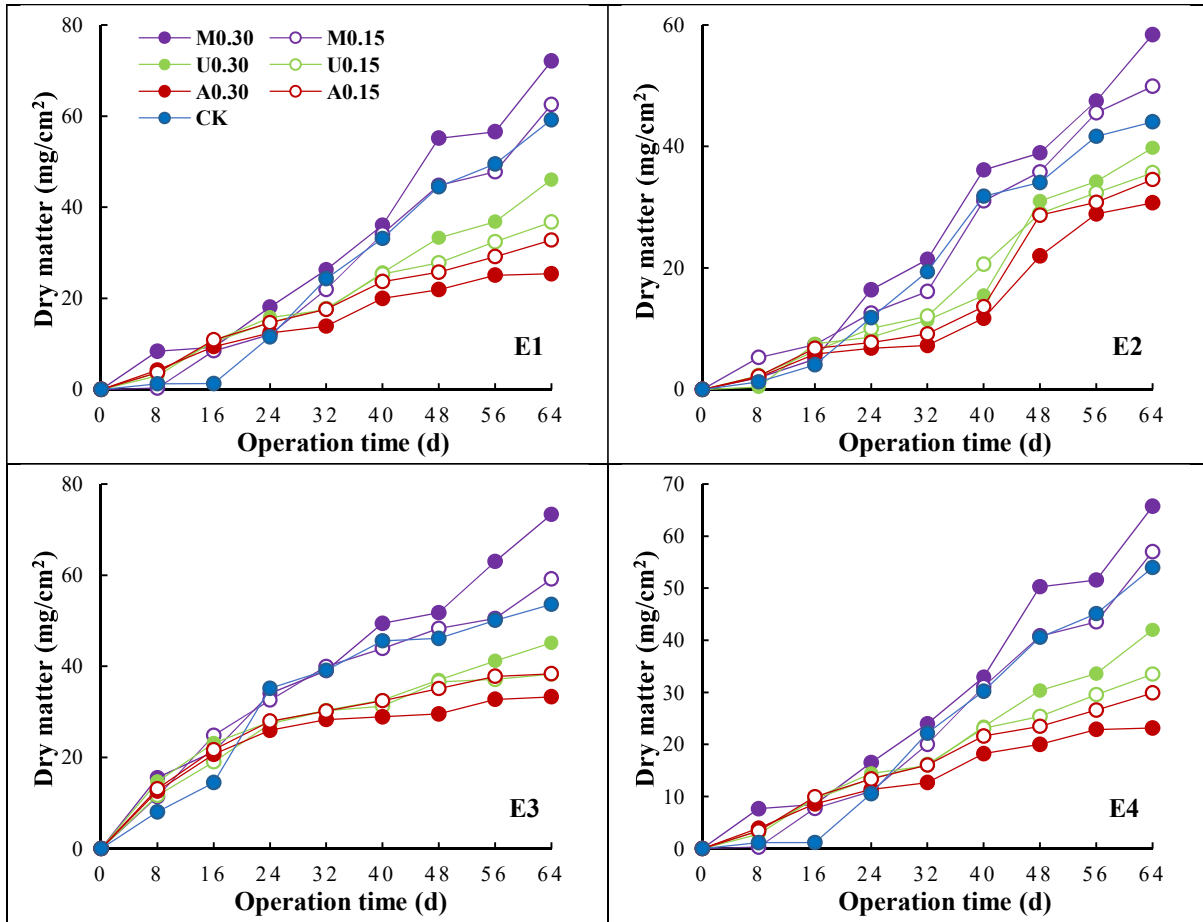


Fig. A5. Dynamic variation of dry matter weight for the different emitters and fertilization treatments.

Table A4. Paired samples t-test of emitter clogging DW in different treatments.

| Treatments | CK | MKP_0.15 | UP_0.15 | APP_0.15 | MKP_0.30 | UP_0.30 | APP_0.30 |
|---|---|---|---|---|---|---|---|
| CK | — | * | ** | ** | ** | ** | ** |
| MKP_0.15 | | — | ** | ** | ** | ** | ** |
| UP_0.15 | | | — | ** | ** | ** | ** |
| APP_0.15 | | | | — | ** | ** | ** |
| MKP_0.30 | | | | | — | ** | ** |
| UP_0.30 | | | | | | — | ** |
| APP_0.30 | | | | | | | — |

Note: asterisks indicate that differences between treatments are significant at $p < 0.05$ (*) or $p < 0.01$ (**).

## 2.4. Basic mineral components of clogging substances found inside emitters (E1-E4).

**(a) MKP_0.30**

Legend:
- Phosphate
- SiO2
- CaCO3-R
- CaCO3-O
- CaMgCO3
- CaCO3·H2O
- CaAl2Si2O8
- Mg5Al(Si3Al)O10(OH)8
- K(Mg,Al)2.04(Si3.34Al0.66)O10(OH)2

**(b) MKP_0.15**

**(c) UP_0.30**

**(d) UP_0.15**

**(e) APP_0.30**

**(f) APP_0.15**

**(g) CK**

E2

**E3**
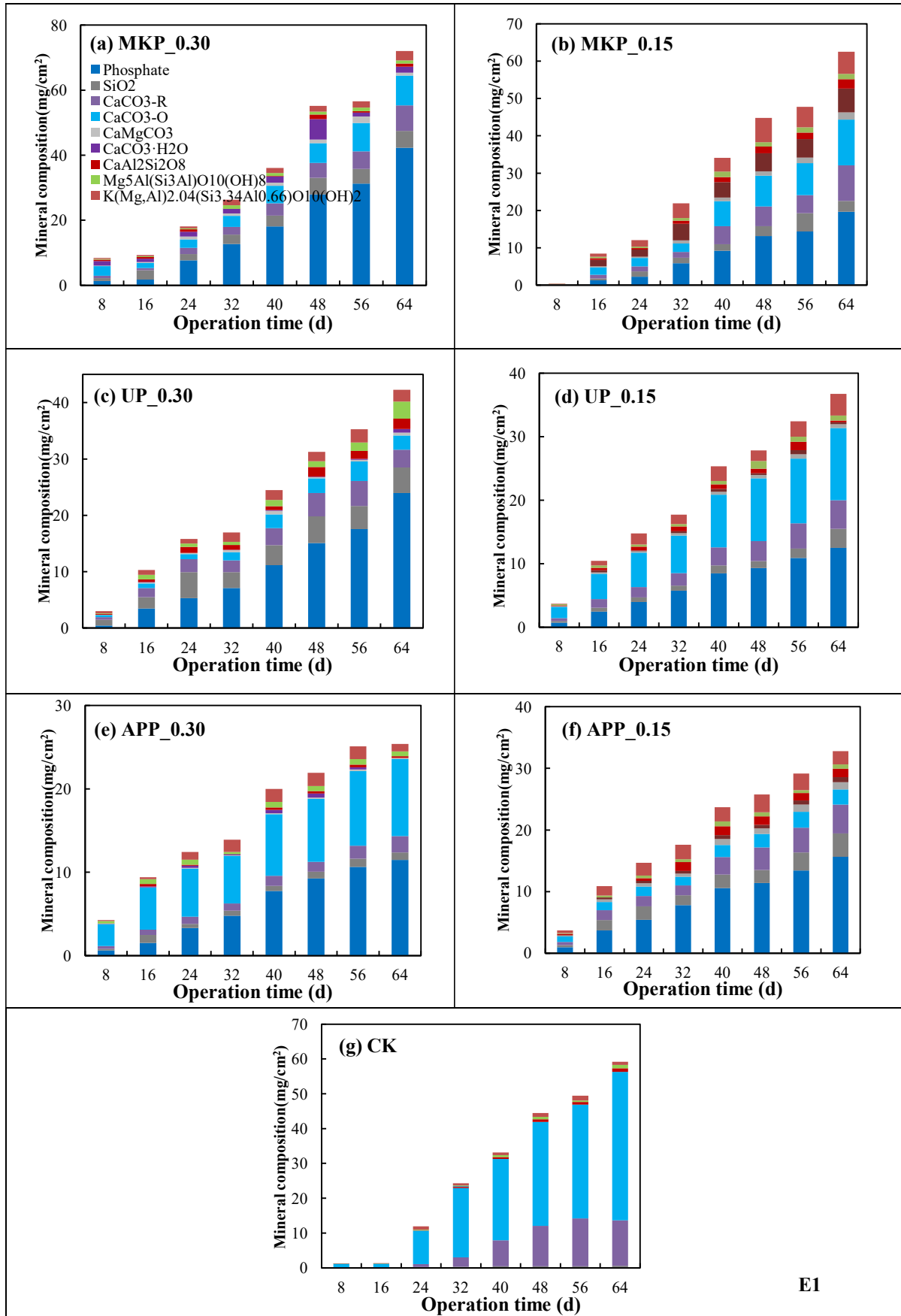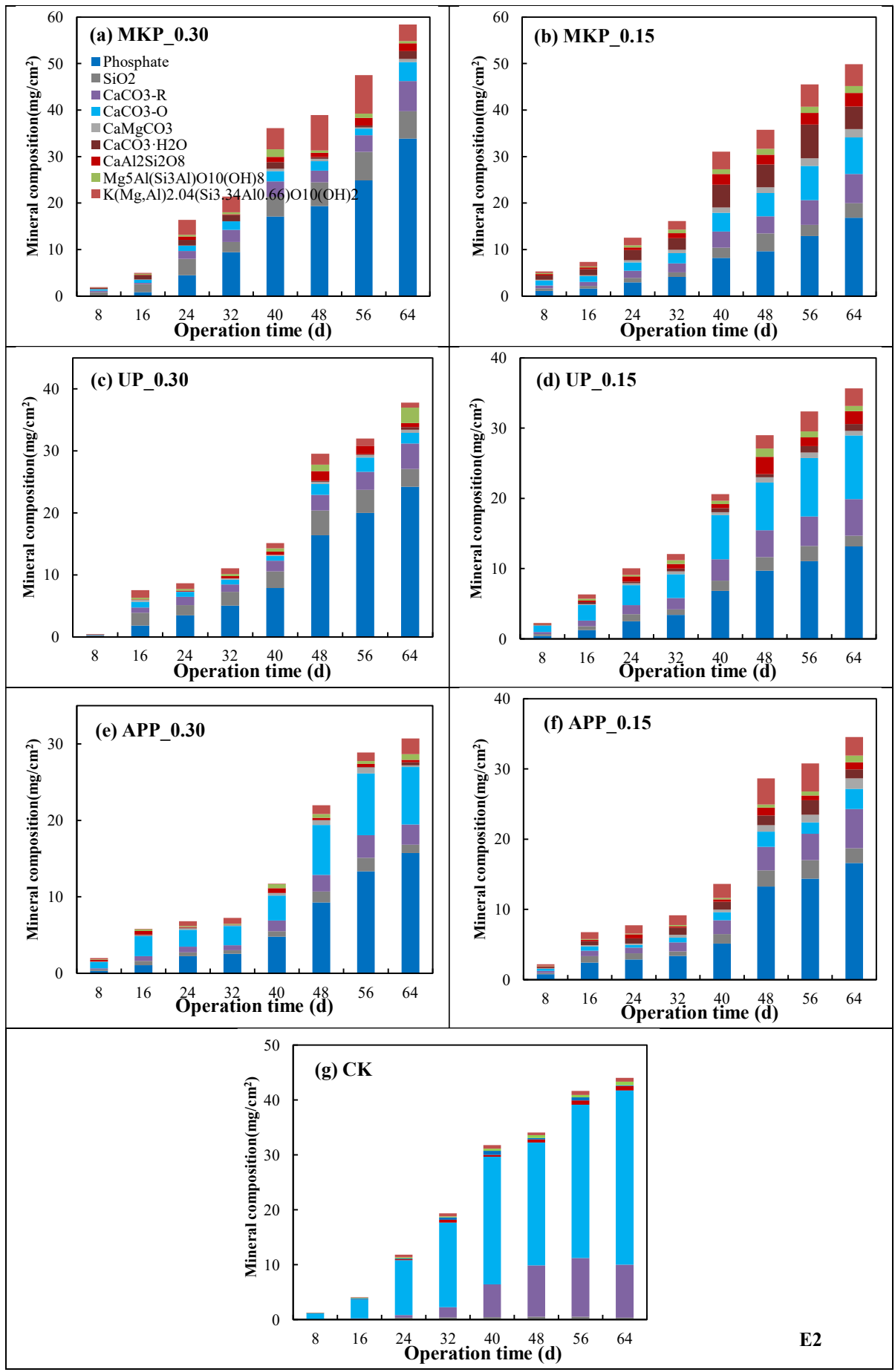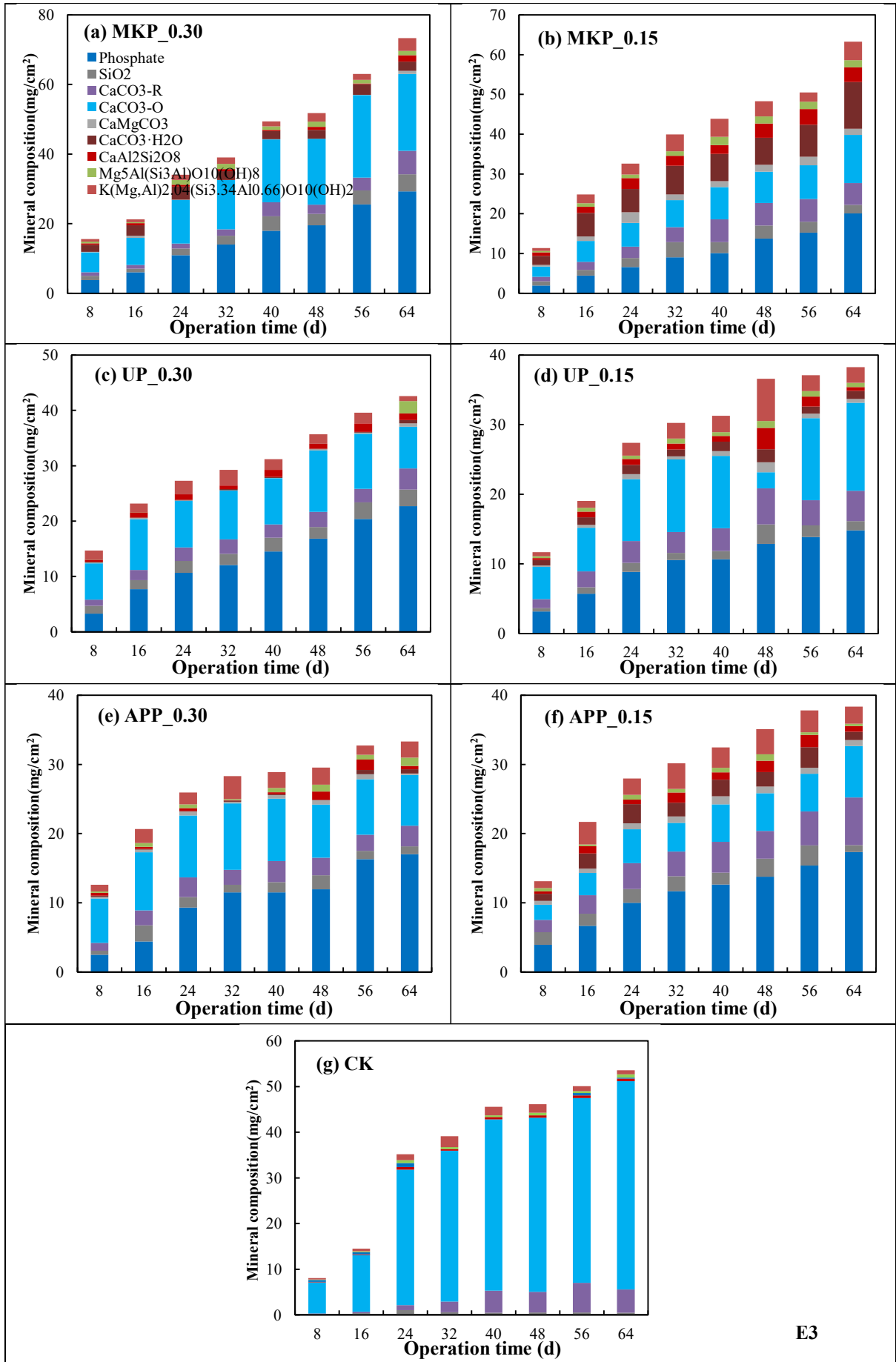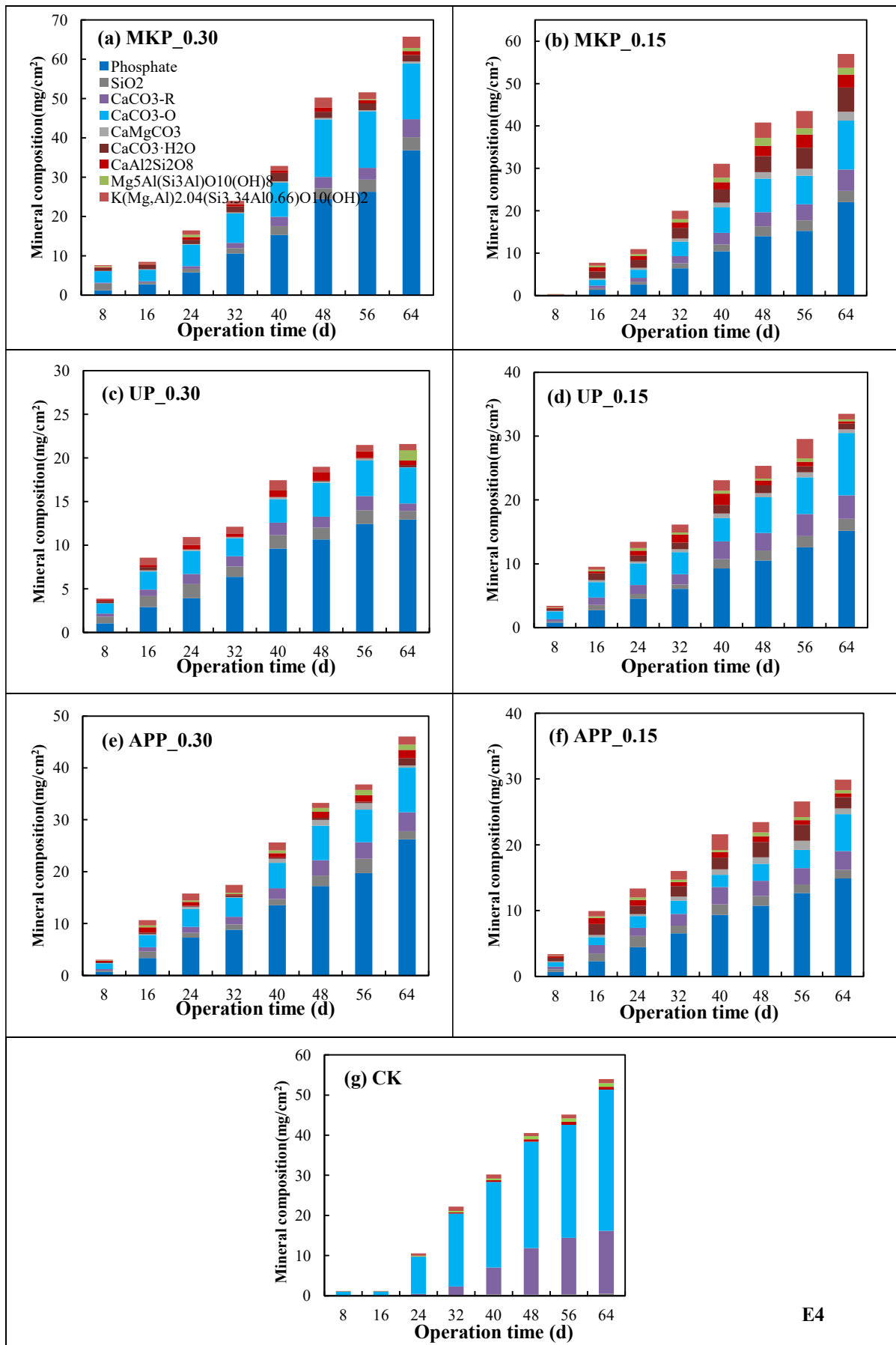
Fig. A6. Dynamic change of the mineral components for the different emitters and fertilizer treatments.

E4

## 2.5. Determination of amorphous composition in MKP, UP and APP treatments

In order to further explore the specific composition of non-crystalline part of clogging substance in emitter under the condition of phosphate fertilizer application in drip irrigation of high salinity water, the component of amorphous components was tested with a Fourier transform infrared spectrometer (FTIR) (manufacturer: PerkinElmer, USA; type: PE Spectrum 400), and scanning electron microscope with an energy Dispersive Spectrometer (SEM-EDS) (manufacturer: Jeol, Japan, type: JSM-6510A). The results of FTIR and SEM-EDS are shown in Fig. A7 and Fig. A8. Fig. A7 indicated that organic substances were not present in amorphous substances. Also, we observed that the microscope views (SEM) across the three types of fertigation treatments varied greatly (Fig. A8), and EDS results (Fig. A8) demonstrated that the clogging substances in the three treatments contained many phosphorus and calcium elements. Thus, the results in the three figures suggested that amorphous components were phosphate.
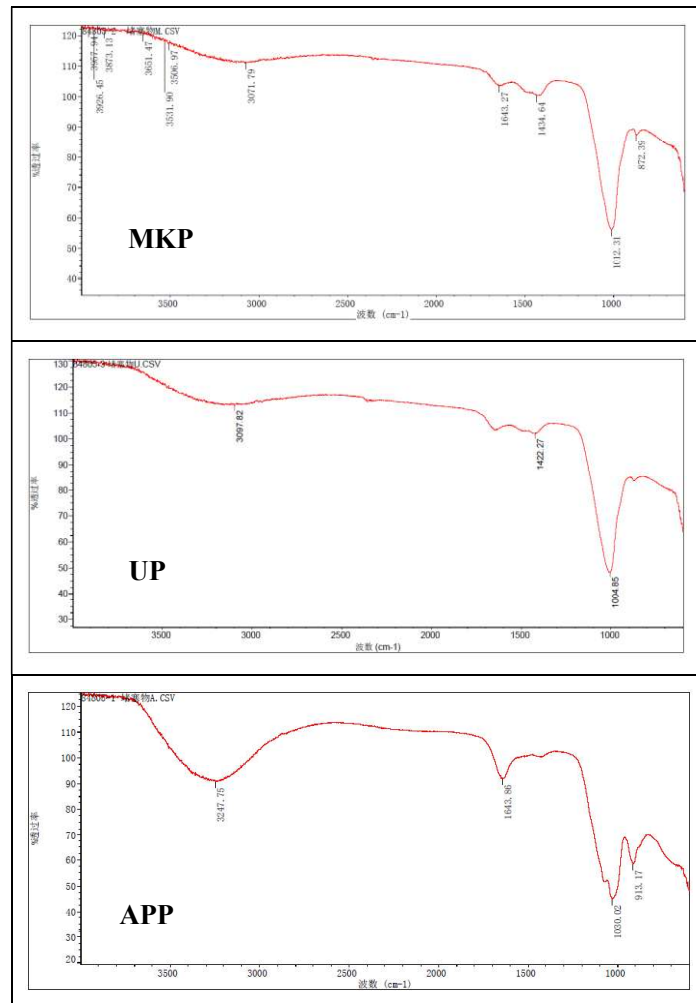


Fig. A7. Infrared spectrum.

## 2.6. Dynamic variation of mineral components
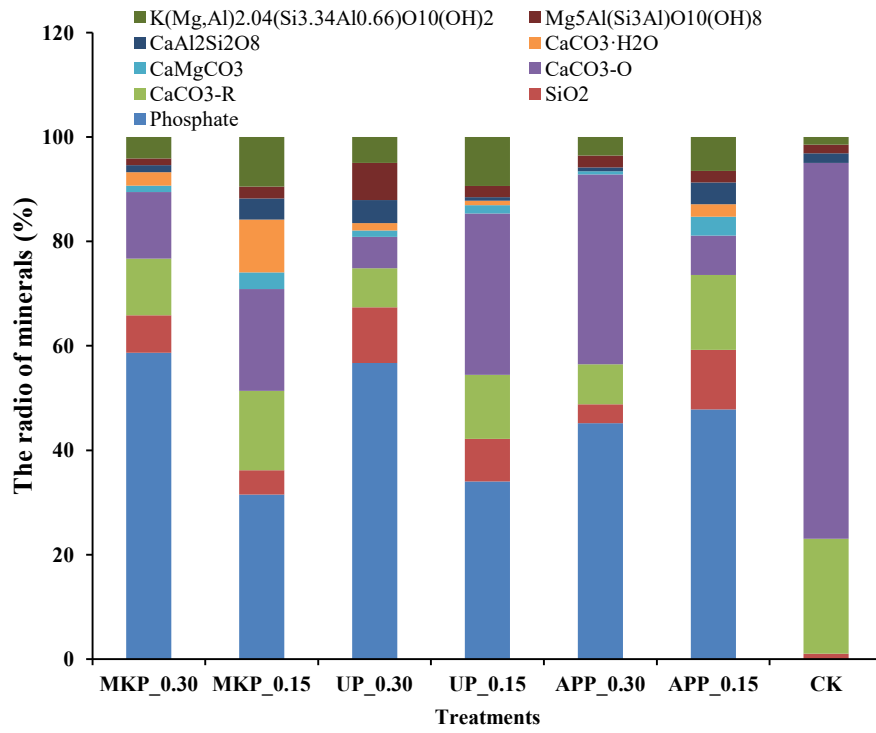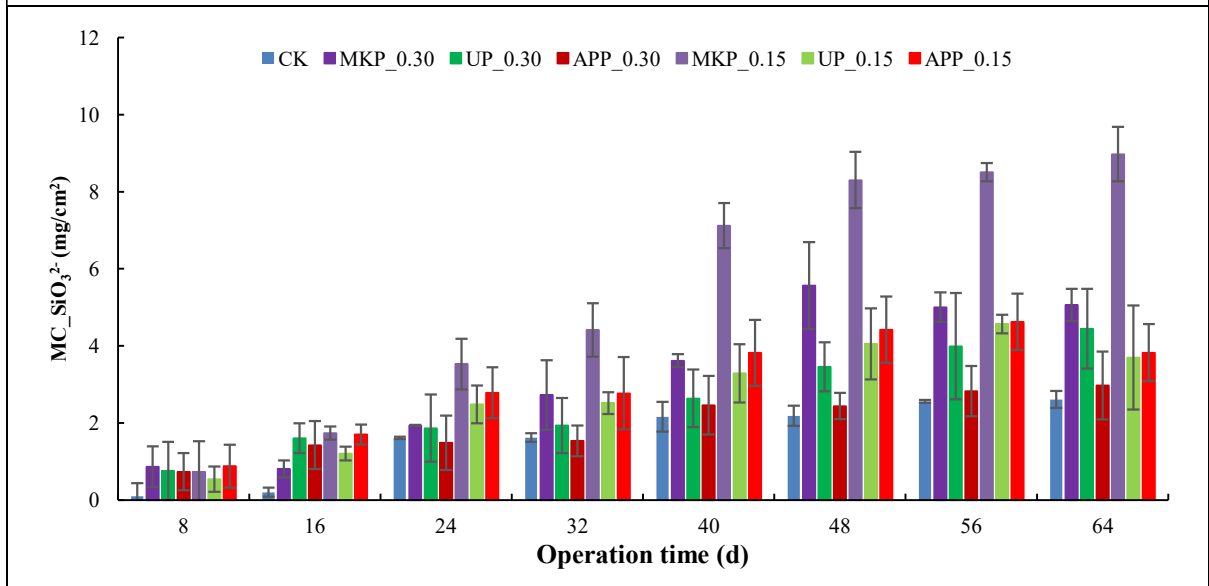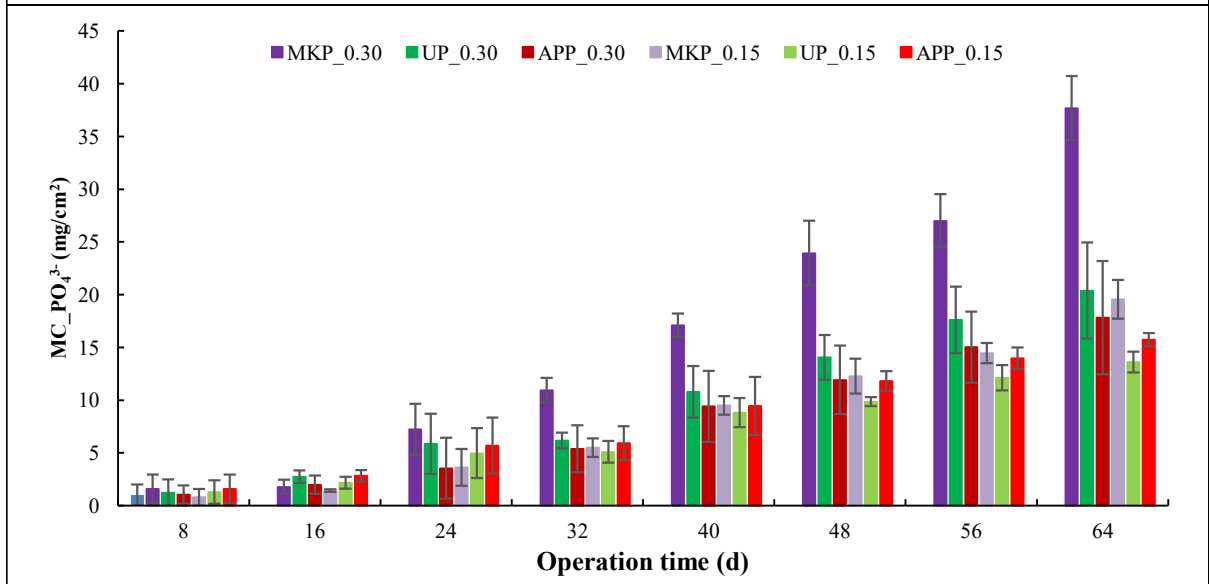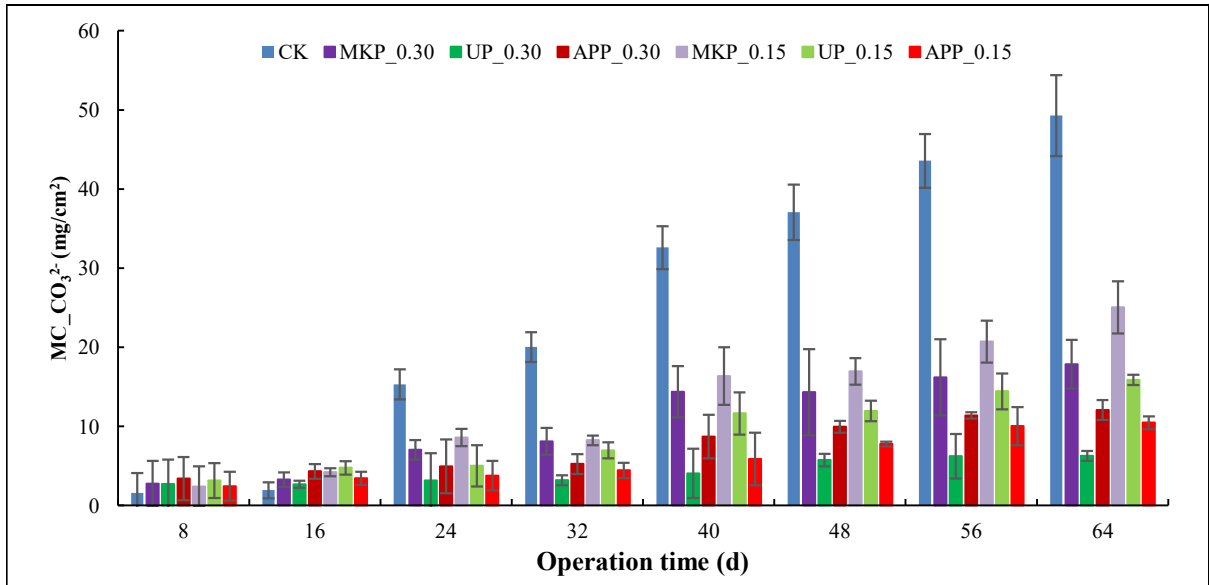


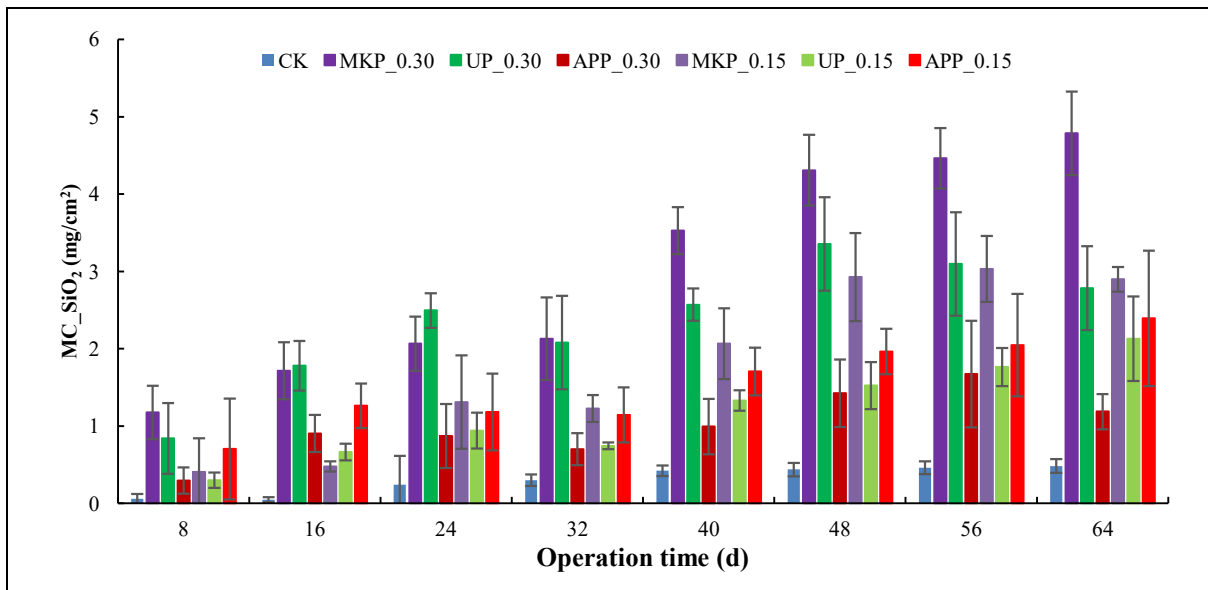Fig. A9. The proportion of mineral components in 7 treatments.

Fig. A10. Changes on average ± standard error of mineral constituent content for the different treatments across the experiment.

Note: there were no phosphate in clogging substances of CK treatment

Table A5. Paired samples t-test of emitter clogging mineral composition in different treatments.

| Minerals | Treatments | CK | MKP_0.15 | UP_0.15 | APP_0.15 | MKP_0.30 | UP_0.30 | APP_0.30 |
|---|---|---|---|---|---|---|---|---|
| Carbonate | CK | — | ** | ** | ** | ** | ** | ** |
| | MKP_0.15 | | — | NS | NS | ** | ** | ** |
| | UP_0.15 | | | — | * | ** | ** | ** |
| | APP_0.15 | | | | — | ** | NS | ** |
| | MKP_0.30 | | | | | — | ** | ** |
| | UP_0.30 | | | | | | — | ** |
| | APP_0.30 | | | | | | | — |
| phosphate | CK | — | ** | ** | ** | ** | ** | ** |
| | MKP_0.15 | | — | ** | ** | ** | ** | ** |
| | UP_0.15 | | | — | ** | ** | ** | ** |
| | APP_0.15 | | | | — | ** | ** | ** |
| | MKP_0.30 | | | | | — | ** | ** |
| | UP_0.30 | | | | | | — | * |
| | APP_0.30 | | | | | | | — |
| Silicate | CK | — | ** | ** | ** | ** | * | * |
| | MKP_0.15 | | — | ** | ** | NS | ** | ** |
| | UP_0.15 | | | — | ** | ** | NS | NS |
| | APP_0.15 | | | | — | ** | NS | NS |
| | MKP_0.30 | | | | | — | ** | ** |
| | UP_0.30 | | | | | | — | NS |
| | APP_0.30 | | | | | | | — |
| Quartz | CK | — | ** | ** | ** | ** | ** | ** |
| | MKP_0.15 | | — | ** | ** | * | NS | NS |
| | UP_0.15 | | | — | * | * | ** | ** |
| | APP_0.15 | | | | — | ** | ** | ** |
| | MKP_0.30 | | | | | — | NS | * |
| | UP_0.30 | | | | | | — | NS |
| | APP_0.30 | | | | | | | — |

Note: asterisks indicate that differences between treatments are significant at p < 0.05 (*) or p < 0.01 (**), and NS means not significant at p < 0.05.
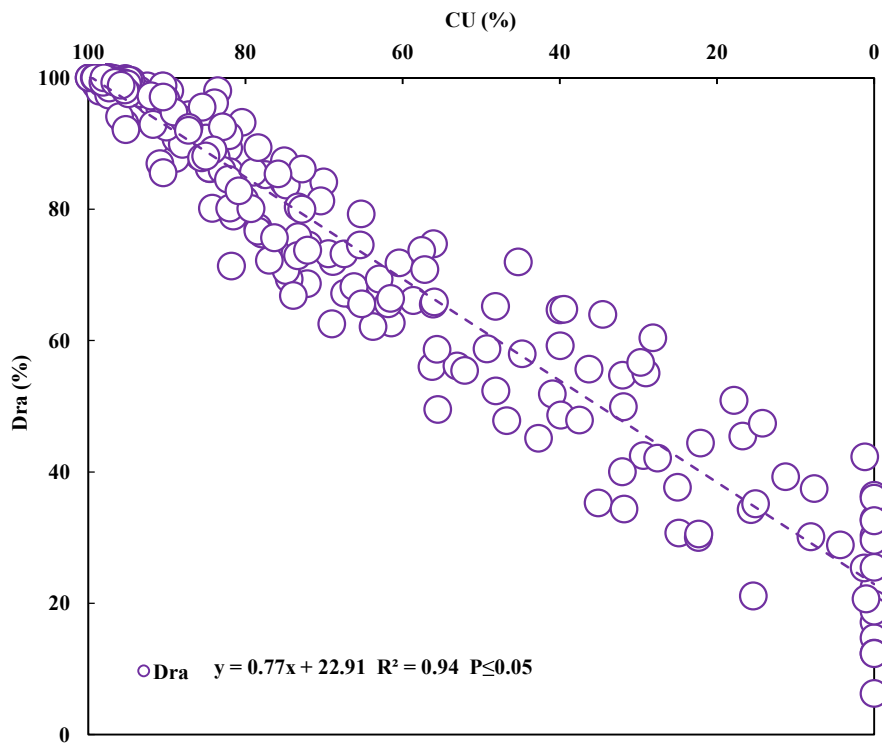
## 2.7. Correlations between *CU* and *Dra*



Fig. A11. Correlations between *CU* and *Dra*.

# 3. Supplementary information in *Discussion section*

## 3.1. Average ± standard deviation of water pH

Table A5. Average ± standard deviation of water pH.

| CK | MKP_0.15 | UP_0.15 | APP_0.15 | MKP_0.30 | UP_0.30 | APP_0.30 |
|---|---|---|---|---|---|---|
| 7.5±0.4 | 6.7±0.4 | 5.6±0.4 | 7.5±0.4 | 6.5±0.4 | 3.1±0.4 | 7.5±0.4 |

Note: pH was measured by a pH meter (manufacture: Rex, China; type: PHSJ-6L).

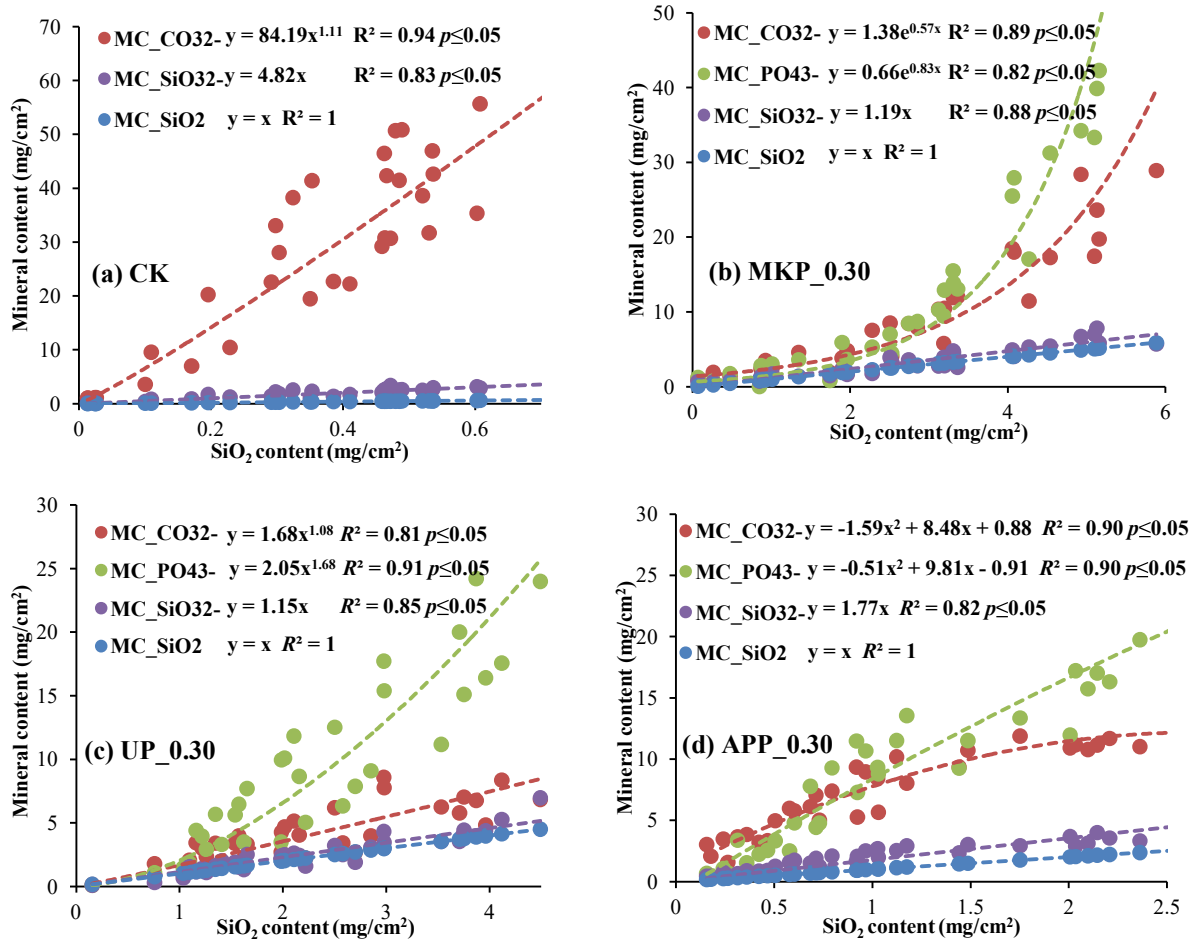## 3.2. Correlation analysis of the content changes of primary mineral components



Fig. A12. Correlation analysis of the content changes of primary mineral components.

Note: (a), (b), (c), and (d) shown the results of CK, MKP_0.30, UP_0.30 and APP_0.30 respectively. The x - axes are the quartz of CK, and the y - axes are the composition of other minerals.