1 **Supplementary Information**

2 **Table S1:** Initial soil chemical properties and basic information about the six long-term experiment sites.

<table>
<thead>
<tr>
<th>Sites</th>
<th>FAO soil classification</th>
<th>Original year</th>
<th>SOC (g·kg⁻¹)</th>
<th>TN (g·kg⁻¹)</th>
<th>AN (mg·kg⁻¹)</th>
<th>TP (mg·kg⁻¹)</th>
<th>AP (mg·kg⁻¹)</th>
<th>TK (g·kg⁻¹)</th>
<th>AK (mg·kg⁻¹)</th>
<th>pH</th>
<th>C/N</th>
<th>Cropping system</th>
</tr>
</thead>
<tbody>
<tr>
<td>GZL</td>
<td>Luvic Phaeozems</td>
<td>1990</td>
<td>13.2</td>
<td>1.40</td>
<td>114.0</td>
<td>1.39</td>
<td>27.0</td>
<td>22.1</td>
<td>190</td>
<td>7.6</td>
<td>9.4</td>
<td>MC-MMM</td>
</tr>
<tr>
<td>ZZ</td>
<td>Calcaric Cambiso</td>
<td>1990</td>
<td>6.7</td>
<td>0.67</td>
<td>76.6</td>
<td>0.65</td>
<td>6.5</td>
<td>16.9</td>
<td>74</td>
<td>8.3</td>
<td>10.0</td>
<td>DC-MW</td>
</tr>
<tr>
<td>UM</td>
<td>Haplic Calcisol</td>
<td>1990</td>
<td>8.8</td>
<td>0.87</td>
<td>55.2</td>
<td>0.67</td>
<td>3.4</td>
<td>19.8</td>
<td>288</td>
<td>8.1</td>
<td>10.1</td>
<td>MC-MWW</td>
</tr>
<tr>
<td>YL</td>
<td>Cumulic Anthrosol</td>
<td>1990</td>
<td>7.4</td>
<td>0.83</td>
<td>63.0</td>
<td>0.61</td>
<td>9.6</td>
<td>21.6</td>
<td>194</td>
<td>8.6</td>
<td>9.0</td>
<td>DC-MW</td>
</tr>
<tr>
<td>ZY</td>
<td>Anthrosol</td>
<td>1982</td>
<td>11.5</td>
<td>0.86</td>
<td>28.1</td>
<td>0.82</td>
<td>21.7</td>
<td>19.8</td>
<td>99</td>
<td>8.5</td>
<td>13.4</td>
<td>MC-MWW</td>
</tr>
<tr>
<td>QY</td>
<td>Eutric Cambisol</td>
<td>1990</td>
<td>7.9</td>
<td>1.07</td>
<td>79.0</td>
<td>0.45</td>
<td>13.9</td>
<td>13.7</td>
<td>104</td>
<td>5.7</td>
<td>7.4</td>
<td>DC-MW</td>
</tr>
</tbody>
</table>

4 GZL, Gongzhuling; ZZ, Zhengzhou; UM, Urumqi; YL, Yangling; ZY, Zhangye; QY, Qiyang; SOC, soil organic carbon; TN, total nitrogen; AN, available nitrogen; TP, total phosphorous; AP, available phosphorous; TK, total potassium; AK, available potassium; C/N, ratio of SOC with TN; MC, monocropping; DC, double-cropping; MMM, maize-maize-maize; MWW, maize-wheat-wheat; MW, maize-wheat.
Table S2: Application rates (kg·ha$^{-1}$) of fertilizer N (inorganic N + N in manure) for each growing season under five treatments.

<table>
<thead>
<tr>
<th>Sites</th>
<th>Experimental Periods</th>
<th>Crops</th>
<th>CK</th>
<th>NPK</th>
<th>NPKM</th>
<th>hNPKM</th>
<th>NPKS</th>
<th>Types of manure</th>
</tr>
</thead>
<tbody>
<tr>
<td>GZL</td>
<td>1990–2018</td>
<td>Maize</td>
<td>0</td>
<td>165</td>
<td>50+115</td>
<td>74+173</td>
<td>123</td>
<td>CM, PM</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0</td>
<td>188</td>
<td>188</td>
<td>282</td>
<td>188</td>
<td>CM, HM</td>
</tr>
<tr>
<td>ZZ</td>
<td>1990–2011</td>
<td>Maize</td>
<td>0</td>
<td>165</td>
<td>50+115</td>
<td>74+173</td>
<td>123</td>
<td>CM, HM</td>
</tr>
<tr>
<td></td>
<td>Wheat</td>
<td>0</td>
<td>242</td>
<td>85+297</td>
<td>152+595</td>
<td>217</td>
<td>GM</td>
<td></td>
</tr>
<tr>
<td>UM</td>
<td>1990–2011</td>
<td>Maize</td>
<td>0</td>
<td>165</td>
<td>50+115</td>
<td>74+173</td>
<td>123</td>
<td>CM</td>
</tr>
<tr>
<td></td>
<td>Wheat</td>
<td>0</td>
<td>242</td>
<td>85+297</td>
<td>152+595</td>
<td>217</td>
<td>GM</td>
<td></td>
</tr>
<tr>
<td>YL</td>
<td>1990–2014</td>
<td>Maize</td>
<td>0</td>
<td>165</td>
<td>50+115</td>
<td>74+173</td>
<td>123</td>
<td>CM</td>
</tr>
<tr>
<td>ZY</td>
<td>1982–1990</td>
<td>Maize</td>
<td>0</td>
<td>240</td>
<td>240+240</td>
<td>NA</td>
<td>NA</td>
<td>PM</td>
</tr>
<tr>
<td></td>
<td>Wheat</td>
<td>0</td>
<td>120</td>
<td>120+120</td>
<td>NA</td>
<td>NA</td>
<td>PM</td>
<td></td>
</tr>
<tr>
<td>ZY</td>
<td>1991–2002</td>
<td>Maize</td>
<td>0</td>
<td>300</td>
<td>300+300</td>
<td>NA</td>
<td>NA</td>
<td>PM</td>
</tr>
<tr>
<td></td>
<td>Wheat</td>
<td>0</td>
<td>150</td>
<td>150+150</td>
<td>NA</td>
<td>NA</td>
<td>PM</td>
<td></td>
</tr>
<tr>
<td>QY</td>
<td>1990–2012</td>
<td>Maize</td>
<td>0</td>
<td>210</td>
<td>63+147</td>
<td>95+221</td>
<td>210</td>
<td>PM</td>
</tr>
<tr>
<td></td>
<td>Wheat</td>
<td>0</td>
<td>90</td>
<td>27+63</td>
<td>41+95</td>
<td>90</td>
<td>PM</td>
<td></td>
</tr>
</tbody>
</table>

GZL, Gongzhuling; ZZ, Zhengzhou; UM, Urumqi; YL, Yangling; ZY, Zhangye; QY, Qiyang. Fertilizer regimes: CK, unfertilized control; NPK, inorganic N, phosphorus and potassium; NPKM, inorganic NPK plus manure; hNPKM, high application rate of NPKM; NPKS, inorganic NPK with stover return; In front and behind of the "+" represents the application rate of N from inorganic fertilizer and manure, respectively; CM, cow manure; PM, pig manure; HM, horse manure; GM, goat manure. NA, data not available because the treatments are not included.
Table S3: Results of principal components analysis (PCA) of nutrient content and soil stoichiometry in the first decade and later period of the experiment.

<table>
<thead>
<tr>
<th>Factors</th>
<th>PC1</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Soil nutrients of the first decade</strong></td>
<td></td>
</tr>
<tr>
<td>soil total nitrogen content (TN, g·kg⁻¹)</td>
<td>-0.776**</td>
</tr>
<tr>
<td>Soil available content (AN, mg·kg⁻¹)</td>
<td>-0.432*</td>
</tr>
<tr>
<td>Soil total phosphorus (TP, g·kg⁻¹)</td>
<td>-0.772**</td>
</tr>
<tr>
<td>Soil available phosphorus (AP, mg·kg⁻¹)</td>
<td>-0.772**</td>
</tr>
<tr>
<td>Soil total potassium (TK, g·kg⁻¹)</td>
<td>0.191</td>
</tr>
<tr>
<td>Soil available potassium (AK, mg·kg⁻¹)</td>
<td>-0.661**</td>
</tr>
<tr>
<td>Cumulative (%)</td>
<td>40.93</td>
</tr>
<tr>
<td><strong>Soil stoichiometry of the first decade</strong></td>
<td></td>
</tr>
<tr>
<td>C/N ratios</td>
<td>-0.474*</td>
</tr>
<tr>
<td>N/P ratios</td>
<td>-0.944**</td>
</tr>
<tr>
<td>C/P ratios</td>
<td>-0.994**</td>
</tr>
<tr>
<td>Cumulative (%)</td>
<td>98.67</td>
</tr>
<tr>
<td><strong>Soil nutrients of the later period</strong></td>
<td></td>
</tr>
<tr>
<td>soil total nitrogen content (TN, g·kg⁻¹)</td>
<td>-0.912**</td>
</tr>
<tr>
<td>Soil available content (AN, mg·kg⁻¹)</td>
<td>-0.870**</td>
</tr>
<tr>
<td>Soil total phosphorus (TP, g·kg⁻¹)</td>
<td>-0.915**</td>
</tr>
<tr>
<td>Soil available phosphorus (AP, mg·kg⁻¹)</td>
<td>-0.936**</td>
</tr>
<tr>
<td>Soil total potassium (TK, g·kg⁻¹)</td>
<td>0.202</td>
</tr>
<tr>
<td>Soil available potassium (AK, mg·kg⁻¹)</td>
<td>-0.827**</td>
</tr>
<tr>
<td>Cumulative (%)</td>
<td>67.11</td>
</tr>
<tr>
<td><strong>Soil stoichiometry of the later period</strong></td>
<td></td>
</tr>
<tr>
<td>C/N ratios</td>
<td>-0.106</td>
</tr>
<tr>
<td>N/P ratios</td>
<td>0.971**</td>
</tr>
<tr>
<td>C/P ratios</td>
<td>0.954**</td>
</tr>
<tr>
<td>Cumulative (%)</td>
<td>62.18</td>
</tr>
</tbody>
</table>

*, significant correlation at P < 0.05. **, significant correlation at P < 0.01.
Table S4: Climatic conditions (annual) in the first decade and later period at the six long-term experimental sites under five treatments.

<table>
<thead>
<tr>
<th>Sites</th>
<th>Years</th>
<th>Mean annual Precipitation (mm)</th>
<th>Mean annual Temperature (°C)</th>
<th>Mean annual Humidity (mm)</th>
<th>Mean annual Evaporation (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GZL</td>
<td>1–10</td>
<td>619.50</td>
<td>7.37</td>
<td>63.70</td>
<td>1235.61</td>
</tr>
<tr>
<td></td>
<td>11–28</td>
<td>544.68</td>
<td>7.23</td>
<td>62.08</td>
<td>1390.25</td>
</tr>
<tr>
<td>ZZ</td>
<td>1–10</td>
<td>617.99</td>
<td>14.62</td>
<td>67.40</td>
<td>1772.89</td>
</tr>
<tr>
<td></td>
<td>11–22</td>
<td>682.58</td>
<td>15.35</td>
<td>61.42</td>
<td>1781.65</td>
</tr>
<tr>
<td>UM</td>
<td>1–10</td>
<td>14.71</td>
<td>14.93</td>
<td>40.80</td>
<td>2458.40</td>
</tr>
<tr>
<td></td>
<td>11–22</td>
<td>15.41</td>
<td>15.72</td>
<td>37.33</td>
<td>2635.45</td>
</tr>
<tr>
<td>YL</td>
<td>1–10</td>
<td>358.06</td>
<td>8.81</td>
<td>55.40</td>
<td>1908.64</td>
</tr>
<tr>
<td></td>
<td>11–25</td>
<td>457.15</td>
<td>10.67</td>
<td>52.33</td>
<td>1977.20</td>
</tr>
<tr>
<td>ZY</td>
<td>1–10</td>
<td>468.85</td>
<td>8.73</td>
<td>65.10</td>
<td>1331.89</td>
</tr>
<tr>
<td></td>
<td>11–21</td>
<td>491.88</td>
<td>9.35</td>
<td>63.64</td>
<td>1497.59</td>
</tr>
<tr>
<td>QY</td>
<td>1–10</td>
<td>1497.05</td>
<td>18.00</td>
<td>78.50</td>
<td>1396.96</td>
</tr>
<tr>
<td></td>
<td>11–23</td>
<td>1373.28</td>
<td>18.25</td>
<td>74.85</td>
<td>1326.50</td>
</tr>
</tbody>
</table>

GZL, Gongzhuling; ZZ, Zhengzhou; UM, Urumqi; YL, Yangling; ZY, Zhangye; QY, Qiyang.
Table S5: Soil pH average value and coefficient variations in the first decade and the later period under five long-term treatments.

<table>
<thead>
<tr>
<th>Sites</th>
<th>Years</th>
<th>CK</th>
<th>NPK</th>
<th>NPKM</th>
<th>hNPKM</th>
<th>NPKS</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>pH</td>
<td>CV</td>
<td>pH</td>
<td>CV</td>
<td>pH</td>
</tr>
<tr>
<td>GZL</td>
<td>1–10</td>
<td>7.60</td>
<td>0.02</td>
<td>6.60</td>
<td>0.05</td>
<td>7.48</td>
</tr>
<tr>
<td></td>
<td>11–28</td>
<td>7.56</td>
<td>0.03</td>
<td>6.20</td>
<td>0.03</td>
<td>7.21</td>
</tr>
<tr>
<td>ZZ</td>
<td>1–10</td>
<td>8.57</td>
<td>0.03</td>
<td>8.42</td>
<td>0.01</td>
<td>8.42</td>
</tr>
<tr>
<td></td>
<td>11–22</td>
<td>8.35</td>
<td>0.03</td>
<td>8.34</td>
<td>0.03</td>
<td>8.27</td>
</tr>
<tr>
<td>UM</td>
<td>1–10</td>
<td>8.10</td>
<td>0.03</td>
<td>8.01</td>
<td>0.04</td>
<td>7.92</td>
</tr>
<tr>
<td></td>
<td>11–22</td>
<td>7.65</td>
<td>0.01</td>
<td>7.56</td>
<td>0.02</td>
<td>7.47</td>
</tr>
<tr>
<td>YL</td>
<td>1–10</td>
<td>8.23</td>
<td>0.03</td>
<td>8.23</td>
<td>0.04</td>
<td>8.10</td>
</tr>
<tr>
<td></td>
<td>11–25</td>
<td>8.38</td>
<td>0.01</td>
<td>8.26</td>
<td>0.02</td>
<td>8.10</td>
</tr>
<tr>
<td>ZY</td>
<td>1–10</td>
<td>8.50</td>
<td>0.00</td>
<td>8.50</td>
<td>0.00</td>
<td>8.50</td>
</tr>
<tr>
<td></td>
<td>11–21</td>
<td>8.50</td>
<td>0.00</td>
<td>8.50</td>
<td>0.00</td>
<td>8.50</td>
</tr>
<tr>
<td>QY</td>
<td>1–10</td>
<td>5.76</td>
<td>0.08</td>
<td>5.01</td>
<td>0.11</td>
<td>6.03</td>
</tr>
<tr>
<td></td>
<td>11–23</td>
<td>5.68</td>
<td>0.03</td>
<td>4.50</td>
<td>0.03</td>
<td>5.82</td>
</tr>
</tbody>
</table>

CV, coefficient of variation. NA, data not available because the treatments are not included.
Table S6: Annual rates of change (ARC, g·kg\(^{-1}\)·yr\(^{-1}\)) of SOC in the first decade and later period under five long-term treatments.

<table>
<thead>
<tr>
<th>Sites</th>
<th>Years</th>
<th>CK</th>
<th>NPK</th>
<th>NPKM</th>
<th>hNPKM</th>
<th>NPKS</th>
</tr>
</thead>
<tbody>
<tr>
<td>GZL</td>
<td>1–10</td>
<td>0.08</td>
<td>0.10</td>
<td>0.16</td>
<td>0.26</td>
<td>0.09*</td>
</tr>
<tr>
<td></td>
<td>11–28</td>
<td>-0.23</td>
<td>-0.38*</td>
<td>0.15</td>
<td>0.29</td>
<td>-0.09</td>
</tr>
<tr>
<td>ZZ</td>
<td>1–10</td>
<td>-0.03</td>
<td>0.05</td>
<td>0.22**</td>
<td>0.28**</td>
<td>0.13*</td>
</tr>
<tr>
<td></td>
<td>11–22</td>
<td>0.10</td>
<td>0.09*</td>
<td>0.23**</td>
<td>0.33**</td>
<td>0.15**</td>
</tr>
<tr>
<td>UM</td>
<td>1–10</td>
<td>-0.12*</td>
<td>-0.04</td>
<td>0.24</td>
<td>0.44**</td>
<td>0.05</td>
</tr>
<tr>
<td></td>
<td>11–22</td>
<td>0.58**</td>
<td>0.66*</td>
<td>1.27*</td>
<td>2.16**</td>
<td>0.49*</td>
</tr>
<tr>
<td>YL</td>
<td>1–10</td>
<td>0.19*</td>
<td>0.29**</td>
<td>0.67**</td>
<td>1.03*</td>
<td>0.29*</td>
</tr>
<tr>
<td></td>
<td>11–25</td>
<td>0.09*</td>
<td>0.20</td>
<td>0.24</td>
<td>0.00</td>
<td>0.31**</td>
</tr>
<tr>
<td>ZY</td>
<td>1–10</td>
<td>-0.19</td>
<td>-0.05</td>
<td>0.00</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td>11–21</td>
<td>-0.23</td>
<td>-0.19</td>
<td>0.03</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>QY</td>
<td>1–10</td>
<td>0.05</td>
<td>0.27*</td>
<td>0.62**</td>
<td>1.02**</td>
<td>0.31*</td>
</tr>
<tr>
<td></td>
<td>11–23</td>
<td>-0.07</td>
<td>-0.12</td>
<td>-0.18</td>
<td>-0.29</td>
<td>-0.13</td>
</tr>
</tbody>
</table>

NA, data not available because the treatments are not included; *, significant correlation at P < 0.05. **, significant correlation at P < 0.01.