

Supporting Information

Tracing the fate of phosphorus fertilizer derived cadmium in soil-fertilizer-wheat systems using enriched stable isotope labeling

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SI-1 Additional information on the soils

Long term field trials

The soils in OEN received P fertilization through a triple superphosphate fertilizer that was applied according to Swiss fertilization guidelines (Flisch et al., 2009). The P fertilizer inputs for different crops in the crop rotation scheme and the different fertilizer treatments are described in detail in Hirte et al. (2021). All other nutrients were also applied according to the fertilization guidelines (Flisch et al., 2009; Hirte et al., 2021).

The “N₂P₂K₂Mg” treatment of ZOFE (ZOFE_{NPK}) received 56 and 139 kg N ha⁻¹ (before and after 1981, respectively), 61 and 38 kg P ha⁻¹, 318 and 167 kg K ha⁻¹ and 12 and 6 kg Mg ha⁻¹ (before and after 1991 for P, K and Mg, respectively) (Oberholzer et al., 2014).

Gallet et al. (2003) and Hirte et al. (2021) describe the OEN experimental designs in more detail. Walther et al. (2001), Leifeld and Mayer (2015), and Annaheim et al. (2015) provide greater detail for the ZOFE experimental designs. No information was available on Cd concentrations of the fertilizers that were applied in the long-term field experiments.

Soil sampling

All soils were sampled immediately after the harvest of spring barley in July 2016 in ZOFE and of maize in November 2016 in OEN. Five samples were extracted per treatment plot replicate. A total of 20 samples were extracted per treatment in OEN and 25 in ZOFE. A steel auger was used to collect samples using a plough layer depth of 20 cm along a W-shaped track. For each plot, no samples were taken from the area within a <1 m distance to the plot boundaries to avoid

35 contamination from neighboring plots. The samples were subsequently sieved to <5 mm, mixed
36 into a composite sample for each treatment, air-dried, and stored in plastic boxes at room
37 temperature and ambient humidity until further use.

38

39 **SI-2 Labeling of the fertilizer with a ^{111}Cd spike**

40 A commercial triple superphosphate fertilizer containing 201 g P kg⁻¹ and a Cd concentration of
41 4.93±0.03 mg kg⁻¹ was ground to a fine powder and suspended in deionized water under
42 permanent stirring. A ^{111}Cd spike solution was prepared by dissolving enriched ^{111}Cd at a specific
43 ^{111}Cd abundance of 95.2% (Trace Sciences International Inc., Pilot Point, TX, US) in few drops of
44 concentrated HNO₃ and then diluted to 1 L with deionized water. ^{111}Cd was chosen as tracer
45 isotope due to the absence of isobaric interferences (Rehkämper et al., 2012) and because it is
46 comparably cheap compared to less abundant isotopes (Larner and Rehkämper, 2012). Although
47 the natural abundance of ^{111}Cd is relatively high at 12.8% (Berglund and Wieser, 2011), it still
48 enables a sufficient resolution of the isotope ratios in the soil-fertilizer-wheat systems because the
49 fertilizer is directly labeled (Dürr-Auster et al., 2019). The spike solution was added to the
50 suspended fertilizer and homogenized for 20 minutes under permanent stirring. The slurry was
51 subsequently dried on a heating plate and ground into a fine powder using a ceramic mortar.

52

53 **SI-3 Comparison of the original and labeled fertilizer**

54 The characterization of the labeled fertilizer was done by including it in the sequential extraction
55 procedure. Furthermore, the water-solubility of Cd and the mineralogy of the original fertilizer and
56 of the labeled one were compared. SI table 1 shows the results of the sequential extraction and the
57 water-soluble fraction.

58

59 **Water solubility of Cd in the original fertilizer and in the labeled fertilizer**

60 Water extracts were prepared using deionized water (>18.2 MΩ) and a fertilizer to water (m:v) ratio
61 of 1:10. The suspension was shaken for 2 h on an orbital shaker at 125 rpm and centrifuged for 5
62 min at 2,500 rpm. The supernatant was filtered using a 0.45 μm syringe filter. The water-soluble
63 Cd concentrations amounted to 55% and 45% of the pseudo-total Cd concentration in the original
64 fertilizer and the labeled one, respectively. This finding indicates that the fertilizer mineralogy may
65 have been changed in the evaporation process.

66

67 **Fertilizer mineralogy**

68 The mineralogy of the original fertilizer and the labeled fertilizer were analyzed using Powder X-ray
69 Diffraction (X'Pert³ Powder, Malvern Panalytical). A semi-quantitative analysis using Griceite as the

70 internal standard revealed that the original fertilizer consisted of about 68% crystalline and 32%
 71 amorphous compounds, whereas the labeled fertilizer consisted of about 36% crystalline and 64%
 72 amorphous compounds. While monocalcium phosphate monohydrate ($\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot (\text{H}_2\text{O})$)
 73 constituted about 59% of the original fertilizer, it only accounted for about 9% of the labeled
 74 fertilizer. Furthermore, the labeled fertilizer consisted of about 19% dicalcium phosphate anhydrous
 75 (CaHPO_4 , monetite). Monetite is formed when H_2O is liberated from dicalcium phosphate dihydrate
 76 ($\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$, brushite) when heated over 80 °C (Dorozhkin, 2007). It is reasonable to expect
 77 this temperature at the interface of the heating plate. Nonetheless, monetite is considered largely
 78 isotopically exchangeable and easily plant-available in soils (Frossard et al., 2004; Lombi et al.,
 79 2006; McDowell et al., 2002; Mengel et al., 2001). It was therefore assumed that the altered P and
 80 Cd speciation in the labeled fertilizer would not strongly influence the plant uptake of the ^{111}Cd
 81 spike. The labeled fertilizer was therefore considered to be a model fertilizer and not equivalent to
 82 the commercial triple superphosphate fertilizer.

83

84 $^{111}\text{Cd}/^{110}\text{Cd}$ isotope ratios in the labeled fertilizer

85 The $^{111}\text{Cd}/^{110}\text{Cd}$ isotope ratios in the four fractions determined through the sequential extraction
 86 procedure and the water extracts show the homogenous distribution of the ^{111}Cd spike in the
 87 labeled fertilizer (SI table 1). A Kruskal-Wallis rank sum test showed no significant differences
 88 between the ratios ($\chi^2=6.267$, $df=4$, $p=0.18$).

89

90 **SI table 1**

91 Characteristics of the labeled fertilizer. Values are presented as means and
 92 one standard deviation (SD) of n=3 samples.

Fractions of the labeled fertilizer	$^{111}\text{Cd}/^{110}\text{Cd}$		Cd concentration		Cd partitioning*	
	[-]	SD	$\mu\text{g g}^{-1}$	SD	%	SD
F1	58.4	± 0.9	25.5	± 1.2	50.1	± 2.3
F2	58.7	± 0.3	22.1	± 0.8	43.4	± 1.6
F3	55.8	± 1.2	2.0	± 0.0	3.8	± 0.1
F4	57.7	± 2.9	1.4	± 0.1	2.8	± 0.1
ΣF1-F4	-		50.9	± 1.5	-	
H₂O	59.1	± 0.7	23.1	± 0.4	45.3	± 0.8

93

94 * Partitioning relative to the summed-up Cd concentration in F1–F4.

95

96 **SI-4 Additional information on the sequential extraction procedure**

97 The revised BCR sequential extraction scheme was chosen because it is standardized and
 98 harmonized in inter-laboratory exercises (Hlavay et al., 2004; Rauret et al., 1999) and because it
 99 consists of fewer extraction steps than other commonly used sequential extraction schemes
 100 (Tessier et al., 1979; Zeien and Brümmer, 1989). Focusing on four fractions reduces the workload

101 and allowed for the analysis of Cd concentrations and isotope ratios in n=4 experimental replicates
102 and for performing robust statistical tests. The sample weight of 1 g, specified in the original
103 method (Rauret et al., 1999), was halved due to the capacity restrictions of the lab equipment.
104 Several studies demonstrated that adaptations of sample weights provide the same results if the
105 sample weight to reagent volume ratio remains unchanged (Bacon et al., 2006; Kazi et al., 2006;
106 Svete et al., 2001).

107 After each decantation, the soil residues were washed by adding 10 mL deionized water (Rauret et
108 al., 1999), shaking for 15 min and centrifuging for 20 minutes at 3000 g. The supernatants of the
109 washing steps were added to the previous extracts. Given that the revised BCR method does not
110 request filtration, it is likely that the supernatants also included colloids.

111 *Aqua regia* digestions for F4 and pseudo-total digestion were performed through stepwise heating
112 until reaching 95 °C in a microwave oven (MARS 6, CEM, Matthews, NC, US) and by maintaining
113 the final temperature for 30 min. The extracts were subsequently filtered through ashless Whatman
114 589/2 filters, diluted to 25 mL with deionized water, and stored at 4 °C until further processing.

115 The Cd_{soil} partitioning was calculated by subtracting the concentration of Cd_{dff} recovered in a
116 sample from the respective Cd concentration. Given that the added Cd_{dff} only minimally contributed
117 to the total Cd concentration of a soil sample (table 1 in the main article), even the erratic Cd_{dff}
118 recoveries found in some OEN samples did not have a significant influence on the calculated Cd_{soil}
119 concentrations.

120

121 **SI-5 Determination of Cd concentrations in labeled samples**

122 **Sample purification through anion exchange resin chromatography**

123 Anion exchange resin chromatography removes matrix elements from the sample to reduce
124 isobaric and non-spectral interferences during mass spectrometry. This removal of matrix elements
125 means that samples do not need to be strongly diluted for measurements with Q-ICP-MS. Lower
126 dilution allows for a more precise determination of Cd concentrations and $^{111}Cd/^{110}Cd$ isotope ratios
127 in samples with low Cd concentrations and/or low ^{111}Cd enrichment.

128 All purification steps were performed in a metal-free clean room laboratory using Biorad Poly-
129 Prep® columns and PFA vials. Solutions used were of analytical grade or higher.

130

131 **Measuring Cd using Q-ICP-MS**

132 Measurements were performed in the collision cell in low helium mode to reduce interferences.
133 The instrument was tuned to high sensitivity while keeping double charges (mass to charge ratio
134 ($m z^{-1}$) 70/140) and oxide ratios ($m z^{-1}$ 156/140) low (both <1.5%). Argon was used as the carrier

135 gas and rhodium was used as an internal standard to monitor and correct signal drift during
136 measurements. The detailed Q-ICP-MS instrumental settings are shown in SI table 2.

137 **SI table 2**

138 Q-ICP-MS instrumental settings.

Parameter	Setting
Integration time per mass [sec]	3.0 for Cd isotopes 0.3 for ¹⁰⁵ Pd, ¹¹⁵ In, ¹¹⁸ Sn
Replicates	5
Sweeps per replicate	1000
Radio frequency power [W]	1550
Radio frequency matching [V]	1.8
Spray chamber temperature [°C]	2.0
Carrier gas (argon) flow [L/min]	1.15
Nebulizer pump [rps]	0.1
Cell helium flow [mL/min]	1.8

139

140 **Determination of ¹¹¹Cd/¹¹⁰Cd ratios**

141 Interference-corrected signal intensities at the mass to charge ratios ($m z^{-1}$) 110 and 111 were
142 used to determine the ¹¹¹Cd/¹¹⁰Cd ratios in soil extracts and plant parts. The ¹¹¹Cd/¹¹⁰Cd ratios in
143 the natural isotopic abundance samples indicated a mass bias of ~2% compared to the ¹¹¹Cd/¹¹⁰Cd
144 ratio calculated using published natural Cd isotope abundances (Berglund and Wieser, 2011).
145 Mass bias effects were corrected by applying a linear correction function (Begley and Sharp, 1997;
146 Ingle et al., 2003; Stürup et al., 2008) that used the natural isotopic abundance ratios determined
147 in purified CMI7003 CRM samples (Silty Clay Loam, Analytika Ltd., Prague, Czech Republic) (SI
148 table 3).

149

150 **Determination of Cd concentrations in labeled samples**

151 The use of ¹¹¹Cd as a tracer isotope made it impossible to determine Cd concentrations by
152 measuring signal intensities at the interference-free $m z^{-1}$ 111. Instead, the signal intensities of all
153 stable Cd isotopes, except ¹¹¹Cd, were calculated using published natural Cd isotopic abundance
154 ratios (Berglund and Wieser, 2011) that were based on interference-corrected signal intensities at
155 $m z^{-1}$ 110. The calculated signal intensities of ¹⁰⁶Cd, ¹⁰⁸Cd, ¹¹²Cd, ¹¹³Cd, ¹¹⁴Cd, and ¹¹⁶Cd were then
156 added to the interference-corrected ¹¹⁰Cd and ¹¹¹Cd signal intensities, which resulted in the total
157 Cd signal intensity used for calculating Cd concentrations. Interference-correction for the signal
158 intensities of ¹¹⁰Cd was done by subtracting the relative contribution of ¹¹⁰Pd on ¹¹⁰Cd using the
159 natural isotopic abundance ¹⁰⁵Pd/¹¹⁰Pd ratio and signal intensities at $m z^{-1}$ 105. $m z^{-1}$ 110 was
160 chosen due to the almost complete elimination of Pd during ion-exchange chromatography and
161 due to presence of Sn at high background concentrations, which made it inappropriate to use
162 individual Sn interference-correction on ¹¹²Cd, ¹¹⁴Cd and ¹¹⁶Cd. Cd concentrations were calculated
163 from signal intensities by deriving a linear equation with the sum of all of the calculated signal

164 intensities of all stable Cd isotopes as the independent variable and a known Cd concentration in
165 calibration standard solutions (IV-ICPMS-71A, Inorganic Ventures, Christiansburg, VA, US) as the
166 dependent variable. Calculated concentrations were also normalized for drifting internal standard
167 signal intensities.

168 The differences in the relative atomic weights of fresh Cd_{diff} in samples were found to be negligible
169 for Cd concentration calculations in soil and plant samples. The relative atomic weight of Cd in the
170 labeled fertilizer as an end-member, where it had the most strongly altered Cd isotopic
171 composition, was 111.22 u. Therefore, its weight was only 1.19 u (1.06%) lower than the standard
172 atomic weight of Cd of 112.41 u (de Laeter et al., 2003). To exclude the effects of altered isotopic
173 abundances of Cd in the presence of the ¹¹¹Cd spike on calculated Cd concentrations, as reported
174 for isotope dilution mass spectrometry (García Alonso and Rodríguez-González, 2013; Pagliano et
175 al., 2015), pseudo-total extracts were also analyzed. These extracts were analyzed using Graphite
176 Furnace Atomic Absorption Spectrometry (GF-AAS; ZEE nit 700P, Analytik Jena, Jena, DE) as an
177 independent method that is not affected by differing isotopic composition of analyzed Cd. The
178 analytical conditions and instrumental parameters proposed by Rucandio et al. (1999) were
179 applied. On average, the Cd concentrations that were measured using GF-AAS amounted to
180 100.79±0.45% of the calculated concentrations of the certified reference material (CMI7003, Silty
181 Clay Loam, Analytika Ltd., Prague, Czech Republic) samples (n=3) and 101.86±1.37% of the
182 pseudo-totally digested labeled Cd_{soil} samples (n=24).

183

184 **SI-6 Additional information on the isotope mixing model**

185 The homogenous distribution of the ¹¹¹Cd stable isotope spike in the different fractions of the
186 labeled fertilizer (SI-3) enabled the use of ¹¹¹Cd/¹¹⁰Cd isotope ratios for source determination. In
187 the MixSIAR mixing models, no informative priors were set to allow equal a priori likelihood of each
188 soil fraction to contribute Cd in wheat shoots. Discrimination was set to zero given that the effects
189 of Cd isotope fractionation during uptake by and relocation in wheat are too small to be determined
190 using a Q-ICP-MS (Imseng et al., 2019; Stürup et al., 2008; Wiggenhauser et al., 2016). The
191 MixSIAR models were run in the predefined “normal” mode for Markov Chain Monte Carlo
192 simulations. The convergence of the models was assessed using Gelman-Rubin diagnostics and
193 Geweke diagnostic (Stock et al., 2018). All models passed the diagnostic tests.

194

195 **SI-7 Quality control**

196 The quality of the extraction and analysis process was assessed by including the certified
197 reference materials CMI7003 (Silty Clay Loam, Analytika Ltd., Prague, Czech Republic) for soil
198 samples, and ERM-CD281 (Rye Grass, Institute for Reference Materials and Measurements, Geel,

199 Belgium) and SRM 1567b (Wheat Flour, National Institute of Standards and Technology,
200 Gaithersburg, MD, US) for plant parts. Certified reference materials were included in triplicate.

201 The accuracy and effectiveness of the sequential extraction procedure was assessed by
202 comparing the summed-up Cd concentrations in F1–F4 in the CMI7003 samples and the pseudo-
203 total Cd concentration in separate 200 mg CMI7003 samples (Rauret et al., 1999). The recovery of
204 certified pseudo-total Cd concentrations were $87.3\pm 2.1\%$ ($n=3$) in the summed-up fractions and
205 $88.5\pm 0.9\%$ ($n=3$) in the separate pseudo-totally digested samples. Other studies also report similar
206 recoveries for the CMI7003 (Jochum et al., 2005; Myöhänen et al., 2002). Furthermore, the
207 summed-up Cd concentrations in F1–F4 in the arable soil samples from OEN and ZOFE were
208 compared to pseudo-total Cd concentrations in separate 200 mg samples of the same arable soil.
209 The Cd concentrations recovered in the summed-up fractions F1–F4 were $103\pm 4.5\%$ of the
210 pseudo-total Cd concentrations in OEN ($n=16$) and $96.3\pm 1.2\%$ in ZOFE ($n=8$).

211 The recoveries of Cd in the certified plant materials were $84.4\pm 3.6\%$ ($n=4$) for ERM-CD281 and
212 $80.9\pm 2.0\%$ ($n=3$) for SRM 1567b. The recoveries obtained were lower than those of the certified
213 values; however, they were consistent with the findings of similar recoveries in other publications
214 (Jochum et al., 2005; Sastre et al., 2002; Wiggenhauser et al., 2016).

215 The recoveries of the applied Cd_{diff} were assessed using a pseudo-total digestion of 200 mg of soil
216 samples and by comparing the mass of Cd_{diff} recovered with the known mass of applied Cd_{diff} . To
217 correct for Cd_{diff} that was taken up by the plants during the experiment, the Cd_{diff} masses in the
218 plants were added to the Cd_{diff} mass in the pseudo-totally digested soil samples. On average, Cd_{diff}
219 recoveries were $92.2\pm 5.7\%$ for the OEN samples ($n=16$) and $108.8\pm 6.3\%$ for the ZOFE samples
220 ($n=8$). The high recoveries of applied Cd_{diff} , even in only 200 mg of soil, indicates that the labeled
221 fertilizer was homogeneously distributed in the 1 kg of soil in the pots during the experiment.

222 The stability of the $^{111}Cd/^{110}Cd$ isotope ratios during the Q-ICP-MS measurements was determined
223 by repeatedly measuring the $^{111}Cd/^{110}Cd$ isotope ratios at natural abundance in the certified
224 reference materials samples. The relative standard deviation over all measurement cycles for the
225 measured $^{111}Cd/^{110}Cd$ isotope ratios ranged between 0.11% and 0.78%.

226 Every batch included method blanks in triplicate. Mean blank Cd concentrations were subtracted
227 from sample Cd concentrations. Mean blank concentrations for soil sample extracts ranged
228 between 0.2 to 9 ng Cd L⁻¹, i.e., a maximum of 0.05% of sample Cd concentrations.

230 Measured $^{111}\text{Cd}/^{110}\text{Cd}$ ratios in certified reference materials (CRM), four soil fractions (F1–
 231 F4), pseudo-totally digested soil samples (PT) and plant parts (roots, straw, grains). All
 232 ratios were corrected for isobaric interferences and mass bias as described in SI-5.

Sample / Past mineral P treatment	Replicate	F1	F2	F3	F4	PT	Roots	Straw	Grains
		$^{111}\text{Cd}/^{110}\text{Cd}$ ratio							
CRM	1	1.027	1.027	1.024	1.023	1.024	1.023	1.025	1.026
	2	1.030	1.025	1.020	1.025	1.025	1.026	1.024	1.022
	3	1.015	1.021	1.028	1.025	1.024	-	-	1.026
OEN _c	1	2.462	1.871	1.254	1.060	1.475	2.520	2.356	2.072
	2	1.804	1.550	1.209	1.063	1.482	2.023	2.085	2.064
	3	2.007	1.658	1.228	1.065	1.632	2.130	2.090	2.030
	4	1.865	1.543	1.203	1.059	1.499	2.036	1.922	2.064
OEN _{1/3}	1	2.315	1.696	1.250	1.066	1.494	2.134	2.086	2.047
	2	1.803	1.538	1.212	1.062	1.520	2.036	2.011	2.077
	3	2.704	2.019	1.269	1.068	1.504	2.035	2.102	2.057
	4	1.816	1.501	1.211	1.057	1.557	2.048	2.104	2.135
OEN _{3/3}	1	1.743	1.513	1.225	1.049	1.499	2.149	2.173	1.969
	2	2.015	1.626	1.245	1.067	1.413	1.987	1.988	1.977
	3	2.226	1.822	1.241	1.064	1.444	1.991	2.007	1.972
	4	1.871	1.568	1.239	1.070	1.507	2.089	2.005	2.006
OEN _{5/3}	1	1.766	1.496	1.176	1.051	1.426	2.016	2.046	1.944
	2	1.880	1.545	1.191	1.053	1.435	1.959	1.862	1.938
	3	2.158	1.695	1.229	1.059	1.448	1.976	1.936	1.946
	4	1.811	1.433	1.207	1.058	1.471	2.022	1.896	1.900
ZOFEC	1	2.666	1.790	1.238	1.093	1.813	2.225	2.118	2.247
	2	2.133	1.619	1.227	1.081	1.861	2.223	2.266	2.246
	3	2.176	1.635	1.226	1.087	1.871	2.261	2.237	2.284
	4	2.152	1.620	1.226	1.092	1.811	2.292	2.289	2.323
ZOFENPK	1	2.350	1.776	1.278	1.073	2.226	2.530	2.573	2.575
	2	2.446	1.838	1.288	1.096	1.926	2.592	2.629	2.610
	3	2.384	1.774	1.254	1.077	1.982	2.544	2.634	2.604
	4	2.423	1.815	1.271	1.077	2.071	2.515	2.358	2.559

234 **SI table 4**

235 Dry matter production, Cd concentrations, total Cd uptake, Cd_{diff} uptake, and Cd_{diff} recovery in plant
 236 parts. Values are presented as medians and as the median absolute deviation (MAD) of n=4
 237 samples. Different letters indicate significant differences (p<0.05) of the medians determined using
 238 the Kruskal–Wallis test.

Treatment	dry matter		Cd conc.		Cd uptake		Cd _{diff} uptake		Cd _{diff} recovery		
	g	MAD	ng g ⁻¹	MAD	µg	MAD	µg	MAD	%	MAD	
grains											
OEN	c	8.5	0.1 ac	42.3	1.7 b	0.37	0.01 ab	0.05	0.00 c	0.19	0.01 c
	1/3	8.7	0.1 abc	30.6	1.3 a	0.26	0.01 a	0.03	0.00 a	0.14	0.01 a
	3/3	8.9	0.1 ab	36.7	2.0 ab	0.33	0.02 ab	0.04	0.00 ab	0.16	0.01 ab
	5/3	9.2	0.4 b	44.6	1.7 b	0.41	0.02 b	0.05	0.00 bc	0.19	0.01 bc
ZOFE	c	6.5	0.2 d	64.1	2.7 c	0.41	0.02 b	0.06	0.00 d	0.25	0.00 d
	NPK	8.4	0.1 cd	40.8	1.9 b	0.35	0.03 ab	0.07	0.00 d	0.26	0.02 d
straw											
OEN	c	9.6	0.1 ac	67.4	0.6 a	0.62	0.02 a	0.08	0.00 a	0.31	0.01 a
	1/3	10.5	0.1 abc	60.1	2.9 a	0.64	0.03 a	0.09	0.01 a	0.33	0.03 a
	3/3	9.7	0.3 ab	63.1	8.9 a	0.60	0.07 a	0.08	0.01 a	0.29	0.03 a
	5/3	10.7	0.3 b	69.7	2.8 ab	0.76	0.04 b	0.09	0.00 a	0.34	0.02 a
ZOFE	c	10.0	0.2 d	182.6	21.1 c	1.82	0.22 c	0.27	0.03 b	1.05	0.12 b
	NPK	12.3	0.1 cd	102.6	7.0 bc	1.28	0.09 bc	0.23	0.01 b	0.90	0.05 b
roots											
OEN	c	3.3	0.1 ce	269.9	11.8 b	0.89	0.02 bc	0.12	0.01 b	0.47	0.03 b
	1/3	3.5	0.0 ab	211.2	5.5 a	0.73	0.03 a	0.09	0.00 a	0.37	0.02 a
	3/3	3.4	0.0 ac	232.8	7.3 b	0.81	0.05 b	0.10	0.01 ab	0.41	0.02 ab
	5/3	3.6	0.1 bd	277.2	2.4 b	0.98	0.02 c	0.12	0.01 b	0.46	0.02 b
ZOFE	c	3.0	0.1 e	571.3	3.7 c	1.75	0.04 d	0.26	0.00 c	1.03	0.02 c
	NPK	3.8	0.0 d	380.7	16.8 c	1.42	0.08 d	0.26	0.01 c	1.03	0.05 c
shoots											
OEN	c	18.1	0.5 ce	54.3	1.9 bc	0.95	0.02 a	0.13	0.01 a	0.50	0.03 a
	1/3	19.2	0.2 ab	47.3	2.8 a	0.90	0.05 a	0.12	0.01 a	0.47	0.03 a
	3/3	18.6	0.2 ac	52.2	4.5 ab	0.96	0.09 a	0.12	0.01 a	0.47	0.04 a
	5/3	19.9	0.6 bd	57.9	3.3 c	1.15	0.07 b	0.13	0.01 a	0.52	0.03 a
ZOFE	c	16.5	0.3 e	137.7	11.7 e	2.24	0.23 c	0.33	0.03 b	1.30	0.13 b
	NPK	20.7	0.2 d	78.9	5.4 d	1.62	0.14 bc	0.29	0.02 b	1.14	0.09 b
plant											
OEN	c	21.3	0.6 ac	86.2	4.7 b	1.84	0.07 b	0.25	0.01 ab	0.97	0.05 ab
	1/3	22.6	0.3 abc	72.0	3.7 a	1.61	0.07 a	0.21	0.01 a	0.84	0.05 a
	3/3	21.9	0.2 ab	80.6	6.3 b	1.81	0.18 b	0.23	0.02 ab	0.89	0.07 ab
	5/3	23.5	0.7 b	88.8	2.4 b	2.13	0.04 c	0.25	0.01 b	0.98	0.02 b
ZOFE	c	19.5	0.3 d	204.6	11.4 c	3.92	0.27 d	0.60	0.03 c	2.33	0.12 c
	NPK	24.5	0.3 cd	126.2	5.6 c	3.07	0.20 d	0.57	0.02 c	2.22	0.07 c

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240 **Additional cited literature**

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