Readily available phosphorous and nitrogen counteract for arsenic uptake and distribution in wheat (*Triticum aestivum* L.)

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Supplementary Information

Table S1: Selected physical and chemical properties of the two different soils (S1, S2).

Property	**S1**	**S2**
Particle size (%)		
2000-630 µm	1.8 ± 0.2	5.4 ± 0.5
630-200 µm	67.3 ± 1.9	12.5 ± 0.5
200-63 µm	17.4 ± 2.2	12.1 ± 0.6
63-20 µm	4.7 ± 1.7	13.2 ± 1.0
20-6.3 µm	2.5 ± 1.7	10.5 ± 0.5
6.3-2 µm	1.3 ± 1.0	7.7 ± 1.2
< 2 µm	4.9 ± 0.5	38.5 ± 0.6
pH ($CaCl_2$)	5.3 ± 0.1	6.0 ± 0.1
CEC (cmol kg^{-1})	5.4 ± 0.4	29.4 ± 1.3
C (%)	2.2 ± 0.3	3.2 ± 0.1
N (%)	0.11 ± 0.01	0.35 ± 0.01
C/N ratio	20.6 ± 0.7	9.1 ± 0.1
P (mg g^{-1})	0.76 ± 0.04	1.12 ± 0.13
Fe (mg g^{-1})	18.7 ± 0.6	59.1 ± 1.7
Mn (mg g^{-1})	0.33 ± 0.01	0.41 ± 0.02
Cu (µg g^{-1})	47.4 ± 2.0	14.9 ± 0.4
Zn (µg g^{-1})	244 ± 8	58 ± 7
As (µg g^{-1})	26.5 ± 1.5	163.9 ± 12.3

Table S2: Results of the ANOVA tests for the effects of moisture treatment (moist), P- and N-fertilization on total As concentrations in different plant parts.

	grain		leaf		stem		root	
	Expl. Var.	F-Value	Expl. Var.	F-Value	Expl. Var.	F-Value	Expl. Var.	F-Value
	%		%		%		%	
S1 (sand)								
moist	0.2	0.6 ns	3.2	51.5 ***	0.0	0.1 ns	7.7	5.9 *
P	61.9	166.7 ***	38.8	613.7 ***	83.4	359.6 ***	16.4	12.5 **
N	15.1	40.7 ***	43.8	693.4 ***	7.3	31.6 ***	10.0	7.6 **
moist x P	4.5	12.0 **	0.4	5.7 *	2.9	12.4 **	0.0	0.0 ns
moist x N	1.2	3.1 ns	0.8	12.0 **	0.1	0.3 ns	6.7	5.1 *
P x N	0.9	2.5 ns	8.2	129.4 ***	0.9	3.9 ns	12.3	9.4 **
moist x P x N	2.2	5.8 *	0.0	0.4 ns	0.2	0.9 ns	5.7	4.3 *
Sum Expl.Var.	86.0		95.1		94.8		58.7	
S2 (loam)								
moist			12.6	32.8 ***	6.0	5.0 *	30.8	30.4 ***
P			25.5	66.5 ***	37.9	32.0 ***	11.4	11.2 **
N			32.6	85.0 ***	4.5	3.8 ns	1.7	1.6 ns
moist x P			11.2	29.2 ***	6.4	5.4 *	20.7	20.4 ***
moist x N			0.0	0.0 ns	0.0	0.0 ns	1.4	1.4 ns
P x N			2.5	6.4 *	1.1	0.9 ns	0.2	0.2 ns
moist x P x N			1.3	3.4 ns	6.2	5.2 *	1.0	1.0 ns
Sum Expl.Var.			85.6		62.1		67.2	

Explained Variation (Expl.Var.) in percent and F-values with significance level (ns = not significant; *: $p<0.05$; **: $p<0.01$; ***: $p<0.001$).

Table S3: As species concentration in leaf and grain including total As and recovery in percent at the time of harvest (plant dieback).

		As(III)	DMA	MMA	As(V)	Total As	Recovery
		mg*kg^{-1}	mg*kg^{-1}	mg*kg^{-1}	mg*kg^{-1}	mg*kg^{-1}	%
S1	**grain**						
	P0_N0_70	0.121 ±0.004	<LOD	<LOD	0.126 ±0.050	0.217 ±0.047	116 ±19
	P1_N0_70	0.317 ±0.048	<LOD	<LOD	0.253 ±0.028	0.646 ±0.113	90 ±12
	P0_N0_100+	0.151 ±0.012	<LOD	<LOD	0.126 ±0.019	0.389 ±0.118	77 ±23
	P1_N0_100+	0.219 ±0.018	<LOD	<LOD	0.214 ±0.019	0.525 ±0.058	83 ±11
	leaf						
	P0_N0_70	0.054 ±0.011	0.009 ±0.002	<LOD	2.99 ±0.335	2.829 ±0.248	108 ±4
	P1_N0_70	0.110 ±0.018	<LOD	0.008 ±0.002	10.647 ±0.927	10.124 ±0.892	106 ±6
	P0_N0_100+	0.051 ±0.008	0.016 ±0.004	<LOD	4.997 ±0.596	4.721 ±0.526	107 ±4
	P1_N0_100+	0.100 ±0.027	0.014 ±0.003	0.010 ±0.002	14.449 ±1.172	12.74 ±1.176	115 ±3
S2	**grain**						
	P0_N0_70	0.025 ±0.007	<LOD	<LOD	0.006 ±0.018	0.177 ±0.042	42 ±1
	P1_N0_70	0.060 ±0.012	<LOD	<LOD	0.032 ±0.022	0.215 ±0.030	47 ±12
	P0_N0_100+	0.044 ±0.006	<LOD	<LOD	0.013 ±0.019	0.171 ±0.040	53 ±16
	P1_N0_100+	0.067 ±0.025	<LOD	<LOD	0.021 ±0.029	0.204 ±0.050	75 ±27
	leaf						
	P0_N0_70	0.023 ±0.003	<LOD	<LOD	0.442 ±0.070	0.941 ±0.581	58 ±19
	P1_N0_70	0.039 ±0.006	<LOD	<LOD	1.213 ±0.005	1.557 ±0.097	81 ±5
	P0_N0_100+	0.024 ±0.005	0.002 ±0.001	<LOD	0.776 ±0.090	1.120 ±0.125	72 ±1
	P1_N0_100+	0.036 ±0.006	0.003 ±0.001	<LOD	1.898 ±0.282	2.353 ±0.294	82 ±3

Edge analysis

The near-edge spectra of As collected for leaves following die-off presented in Figure 3b suggest the presence of two As species with different oxidation states. Linear combination fits of the near edge spectra with the reference spectra available did not deliver a good fit of the spectra (not shown), indicating that the reference compounds were not representative of the local chemical environment of As in the leaves. To determine As oxidation state, we took the first derivative of the edge region of the normalized As K edge spectra, and compared these to the first derivatives of the near-edge spectra of the arsenite (As(III)) and arsenate (As(V)) reference compounds ($NaAs_2O_2$ and Na_2HAsO_4, respectively) and the orpiment (As_2S_3) reference. The comparison is demonstrated in the figure below for the spectrum collected at Spot 3 (see Figure 3b of the main manuscript), which is representative of the various spectra collected. The comparison shows that: (1) the peaks in the first derivatives of the three As reference compounds are well separated, which facilitates the use of the first-derivative spectra to determine As oxidation state; and (2) the derivative of the As K edge spectrum collected at Spot 3 shows two peaks that correspond to the peaks observed in the derivatives of As(III) and As(V), whereas no evidence is found for the presence of significant amounts of As-S species. These findings are consistent with a more oxidizing environment in the dead leaves (relative to fresh leaves) where sulfur ligands have been replaced with oxygen ligands and partial oxidation to As(V) has occurred.

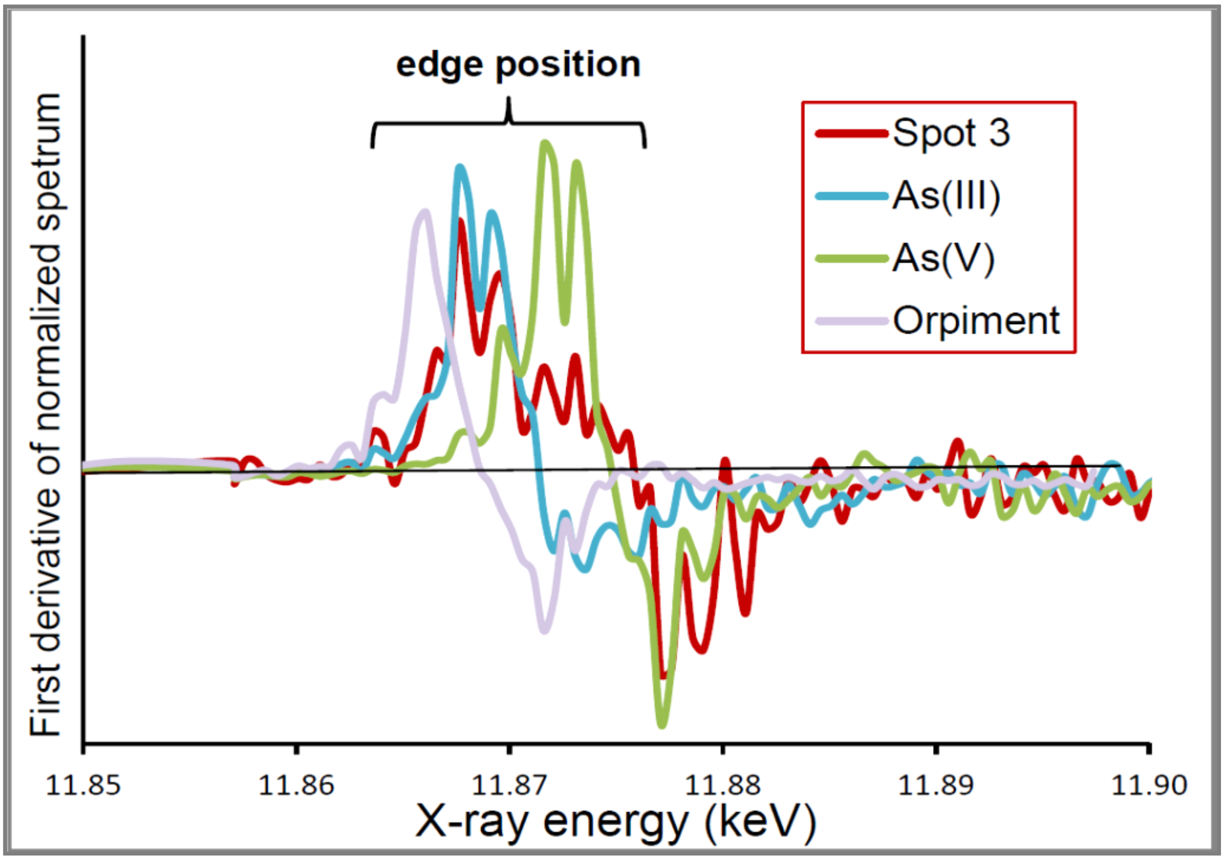

Figure S1. Comparison of the first derivative spectra of the normalized As K edge spectrum collected at Spot 3 in the dead leaf (see Figure 3b of main manuscript) to those of the As(III), As(V) and orpiment reference compounds.

Table S4: As transfer and translocation factors for aboveground plant parts on the sandy soil S1 and on the loamy soil S2 with moisture treatment (70; 100+), P-treatment (P0; P1) and N-treatment (N0; N1); mean with ±sd; n=5. “comp.”= plant part

	S1 (sandy soil)			**S2 (loamy soil)**	
	Transfer factor	Translocation factor		Transfer factor	Translocation factor
grain		comp./root			comp./root
P0_N0_70	0.0085 ±0.0023	0.045 ±0.013		0.0012 ±0.0004	0.007 ±0.002
P1_N0_70	0.029 ±0.0075	0.171 ±0.05		0.0013 ±0.0002	0.02 ±0.008
P0_N1_70	0.0042 ±0.0008	0.033 ±0.024		<LOD	<LOD
P1_N1_70	0.0205 ±0.0036	0.057 ±0.018		<LOD	<LOD
P0_N0_100+	0.0147 ±0.0054	0.094 ±0.053		0.0012 ±0	0.017 ±0.002
P1_N0_100+	0.0199 ±0.0027	0.102 ±0.021		0.0012 ±0.0004	0.02 ±0.003
P0_N1_100+	0.0032 ±0.0027	0.026 ±0.023		<LOD	<LOD
P1_N1_100+	0.0172 ±0.0021	0.074 ±0.02		0.0011 ±0.0003	0.016 ±0.006
leaf					
P0_N0_70	0.1103 ±0.0141	0.591 ±0.141		0.0195 ±0.0297	0.113 ±0.184
P1_N0_70	0.4512 ±0.0757	**2.731 ±0.976**		0.0096 ±0.0005	0.134 ±0.034
P0_N1_70	0.0183 ±0.0051	0.142 ±0.097		0.003 ±0.0018	0.014 ±0.007
P1_N1_70	0.1098 ±0.0168	0.306 ±0.088		0.0022 ±0.0006	0.024 ±0.007
P0_N0_100+	0.1766 ±0.0313	1.15 ±0.641		0.0068 ±0.0008	0.104 ±0.024
P1_N0_100+	0.4869 ±0.078	**2.465 ±0.374**		0.0134 ±0.0021	0.239 ±0.058
P0_N1_100+	0.0263 ±0.0068	0.202 ±0.058		0.0023 ±0.0016	0.066 ±0.068
P1_N1_100+	0.1677 ±0.0294	0.713 ±0.175		0.008 ±0.0005	0.111 ±0.015
stem					
P0_N0_70	0.022 ±0.004	0.118 ±0.036		0.002 ±0.0003	0.012 ±0.005
P1_N0_70	0.1071 ±0.0333	0.627 ±0.199		0.0023 ±0.0003	0.031 ±0.007
P0_N1_70	0.0309 ±0.0057	0.247 ±0.184		0.002 ±0.0003	0.01 ±0.004
P1_N1_70	0.0969 ±0.055	0.266 ±0.184		0.0026 ±0.0005	0.029 ±0.011
P0_N0_100+	0.0347 ±0.0066	0.22 ±0.11		0.0015 ±0.0001	0.024 ±0.004
P1_N0_100+	0.076 ±0.0111	0.385 ±0.06		0.003 ±0.0008	0.054 ±0.016
P0_N1_100+	0.0462 ±0.0058	0.356 ±0.079		0.0021 ±0.0008	0.058 ±0.029
P1_N1_100+	0.1118 ±0.0187	0.487 ±0.173		0.0026 ±0.0002	0.037 ±0.005