SUPPLEMENTAL MATERIAL

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Table S1: ^{15}N-incorporation of the selected peptides of RuBisCO large (L1-3) and small subunit (S1-3) (Table 1) with different permanent ^{15}N-labeling of plants (value ± standard error (SE)); ^{15}N-incorporation was calculated A) with MoLE and B) with the excel sheet from (Taubert et al [19]); L1-3 and S1-3 are numbers of peptides (Table 1).

A

expected ^{15}N labeling [At%]	^{15}N-incorporation ± SE [At%] calculated by MoLE					
	0.37	**1.35**	**5.28**	**10.19**	**49.49**	**98.61**
L1	1.01 ± 0.06	1.93 ± 0.12	5.76 ± 0.09	10.11 ± 0.06	48.88 ± 0.14	99.15 ± 0.06
L2	0.61 ± 0.07	1.46 ± 0.01	5.01 ± 0.06	9.33 ± 0.09	48.15 ± 0.23	99.30 ± 0.09
L3	0.76 ± 0.11	1.74 ± 0.06	5.33 ± 0.10	9.65 ± 0.07	48.35 ± 0.21	99.08 ± 0.07
S1	0.53 ± 0.05	1.42 ± 0.05	5.16 ± 0.08	9.80 ± 0.10	49.03 ± 0.29	99.43 ± 0.08
S2	0.54 ± 0.04	1.47 ± 0.05	5.18 ± 0.03	9.81 ± 0.08	48.91 ± 0.25	98.96 ± 0.44
S3	0.99 ± 0.08	1.94 ± 0.11	5.82 ± 0.09	9.89 ± 0.09	49.39 ± 0.18	99.53 ± 0.21

B

expected ^{15}N labeling [At%]	^{15}N-incorporation ± SE [At%] calculated according to Taubert et al.					
	0.37	**1.35**	**5.28**	**10.19**	**49.49**	**98.61**
L1	0.87 ± 0.06	1.83 ± 0.09	5.47 ± 0.06	10.14 ± 0.04	48.89 ± 0.17	99.12 ± 0.07
L2	0.58 ± 0.06	1.41 ± 0.02	4.86 ± 0.21	9.30 ± 0.09	48.27 ± 0.21	99.07 ± 0.07
L3	0.46 ± 0.06	1.39 ± 0.02	4.91 ± 0.21	9.66 ± 0.09	47.74 ± 0.21	99.06 ± 0.10
S1	0.30 ± 0.03	1.33 ± 0.02	5.21 ± 0.08	9.63 ± 0.07	48.85 ± 0.22	98.95 ± 0.11
S2	0.48 ± 0.07	1.35 ± 0.03	5.17 ± 0.02	9.90 ± 0.09	48.58 ± 0.33	98.92 ± 0.14
S3	1.06 ± 0.06	2.00 ± 0.09	5.49 ± 0.05	10.18 ± 0.06	49.41 ± 0.23	99.00 ± 0.11

Table S2: Peptides of phosphorylase b from rabbit muscle and of BSA (B) used for calculation of the absolute protein quantitation on the column. The three most intense peptides were used. The table includes the average mass and retention time (Rt) calculated from 45-fold analysis. Carbamido methylated methionine residue is denoted as C*.

No.	**Calc.[MH]$^+$**	**Exp. [MH]$^+$**	**Δ ppm***	**Rt [min]**	**Rt RSD [%]**	**Sequence**	**Sumformula**
P1	1853.9644	1853.9734	4.9	50.62	0.5	LLSYVDDEAFIRDVAK	$C_{84}H_{132}N_{20}O_{27}$
P2	1886.9031	1886.9120	4.7	45.46	0.6	GYNAQEYYDRIPELR	$C_{84}H_{123}N_{23}O_{27}$
P3	1678.8646	1678.8759	6.7	46.50	0.6	IGEEYISDLDQLRK	$C_{73}H_{119}N_{19}O_{26}$
B4	1163.6306	1163.6334	2.4	45.96	0.6	LVNELTEFAK	$C_{53}H_{86}N_{12}O_{17}$

B5	1419.6937	1419.7019	5.8	47.64	1.1	SLHTLFGDELC*K	$C_{62}H_{198}N_{16}O_{20}S_1$
B6	1639.9378	1639.9448	4.3	42.62	0.7	KVPQVSTPTLVEV SR	$C_{72}H_{126}N_{20}O_{23}$

Table S3: Peptides of ribulose-1,5-bisphosphate-carboxylase/oxygenases (RuBisCO) LSU (L) and small (S) SSU and RCA2 (R) and lipoxygenase 2 (LO) from *Nicotiana attenuata* and of BSA (B) used for absolute protein quantitation and for calculation of the ^{15}N-incorporation of soil grown plants pulse labeled with $K^{15}NO_3$. The three most intense peptides were taken, except from RuBisCO LSU where the 3th, 6th and 8th most intense peptides were used. The table includes the average mass and retention time (Rt) calculated from 13-fold analysis. Carbamido methylated methionine residue is denoted as C*.

No.	**Calc.[MH]$^+$**	**Exp. [MH]$^+$**	**Δ ppm***	**Rt [min]**	**Rt RSD [%]**	**Sequence**	**Sumformula**
L4	1261.7150	1261.7141	0.7	51.88	0.0	DITLGFVDLLR	$C_{58}H_{96}N_{14}O_{17}$
L5	1261.6285	1261.6303	1.4	42.02	0.4	FLFC*AEALYK	$C_{61}H_{88}N_{12}O_{15}S_1$
L6	1546.7358	1546.7371	0.8	41.65	0.0	WSPELAAAC*EV WK	$C_{71}H_{103}N_{17}O_{20}S_1$
S3	933.5152	933.5159	0.8	34.13	0.0	IIGFDNVR	$C_{42}H_{68}N_{12}O_{12}$
S4	1802.8781	1802.8811	1.7	40.22	0.0	QVQC*ISFIAYKPE GY	$C_{83}H_{123}N_{19}O_{24}S_1$
S5	893.4978	893.4964	1.6	32.85	0.1	EVEYLLK	$C_{42}H_{68}N_8O_{13}$
B7	1305.7161	1305.7172	0.8	31.66	0.0	HLVDEPQNLIK	$C_{58}H_{96}N_{16}O_{18}$
B8	1163.6306	1163.6325	1.6	36.52	0.0	LVNELTEFAK	$C_{53}H_{86}N_{12}O_{17}$
B9	1479.7954	1479.7962	0.5	40.81	0.0	LGEYGFQNALIVR	$C_{68}H_{106}N_{18}O_{19}$
LO1	1142.6051	1142.6052	0.0	35.66	0.0	EALPEDLISR	$C_{49}H_{84}N_{13}O_{18}$
LO2	1572.8631	1572.8665	2.2	47.85	0.0	DVLLFETPELLQR	$C_{72}H_{117}N_{17}O_{22}$
LO3	1629.8370	1629.8397	1.7	34.99	0.0	LDPEIYGPPESAIT K	$C_{74}H_{116}N_{16}O_{25}$
R4	1882.9697	1882.9745	2.5	49.83	0.0	IVDTFPGQSIDFFG ALR	$C_{88}H_{131}N_{21}O_{25}$
R5	1706.7980	1706.8001	1.2	35.78	0.0	GLVQDFSDDQQDI AR	$C_{71}H_{111}N_{21}O_{28}$
R6	1332.6794	1332.6796	0.2	34.05	0.0	WVSGTGIEAIGDK	$C_{59}H_{93}N_{15}O_{20}$

$*\Delta\ ppm=10^6 *(M_{tn}-M_{exp})*M_{tn}^{-1}$

LEGENDS:

Figure S1: Fertilization scheme of a) permanent labeling experiment and b) pulse labeling experiment. a) 12 days after germination plants were transferred to 50 mL single pots with different concentrations of $Ca(^{15}NO_3)_2$ (see Material and Methods). 10 days later they were put into 1 L single pots with the same concentrations of ^{15}N in the form of $K^{15}NO_3$. Ten days later plants were harvested. b) 7 days after transfer to 1 L pots, plants were pulse labeled with $K_{15}NO^3$. Three days later was the first time-point of harvest.

Figure S2: LC-MS^E production spectra of selected peptides (Table 1) a-c) for LSU; d-f) for SSU; g-i) for RCA2; j-l) for BSA2; pe = precursor error

Figure S3: Absolute difference between calculated (excel sheet Taubert et al. [19]) and expected ^{15}N-incorporation at different concentrations of partial permanent ^{15}N-labeling. Mean ± SE (n=5) of three peptides of RuBisCO LSU (L1-3) and SSU (S1-3) (for peptides see Table 1).

Figure 4: Absolute differences of the ^{15}N-incorporation of RuBisCO peptides between technical replicates determined with MoLE from leaf extracts of plants grown at different concentrations of partial permanent ^{15}N-labeling. Mean ± SE (n=5) of the difference between two technical replicates is shown (for peptides see Table 1).

Figure S5: Absolute difference between measured and expected ^{15}N-incorporation of total soluble protein determined by IRMS from leaf extracts of plants grown at different concentrations of partial permanent ^{15}N-labeling. The proteins with an expected ^{15}N-incorporation higher than 5 % were mixed with BSA before analysis to dilute the labeling to about 1 At% ^{15}N-labeling. Mean ± SE (n=5) of the differences is shown.

Figure S6: Absolute differences between technical replicates of the ^{15}N-incorporation in TSP measured with IRMS from leaf extracts of plants grown at different concentrations of partial permanent ^{15}N-labeling. Mean ± SE (n=5) of the difference between two technical replicates is shown. Samples with a labeling higher than 5 % were mixed with BSA before analysis to dilute the labeling to about 1 At% ^{15}N-labeling.

Figure S7: ^{15}N-incorporation of LOX2 protein in leaves of ir*LOX3* plants determined with MoLE and total ^{15}N-incorporation of the same leaf measured by IRMS.Arrows indicate days of treatment. Oldest sink leaves at time point of labeling were harvested at indicated time points. For further details see Fig. 7.

Figure S1

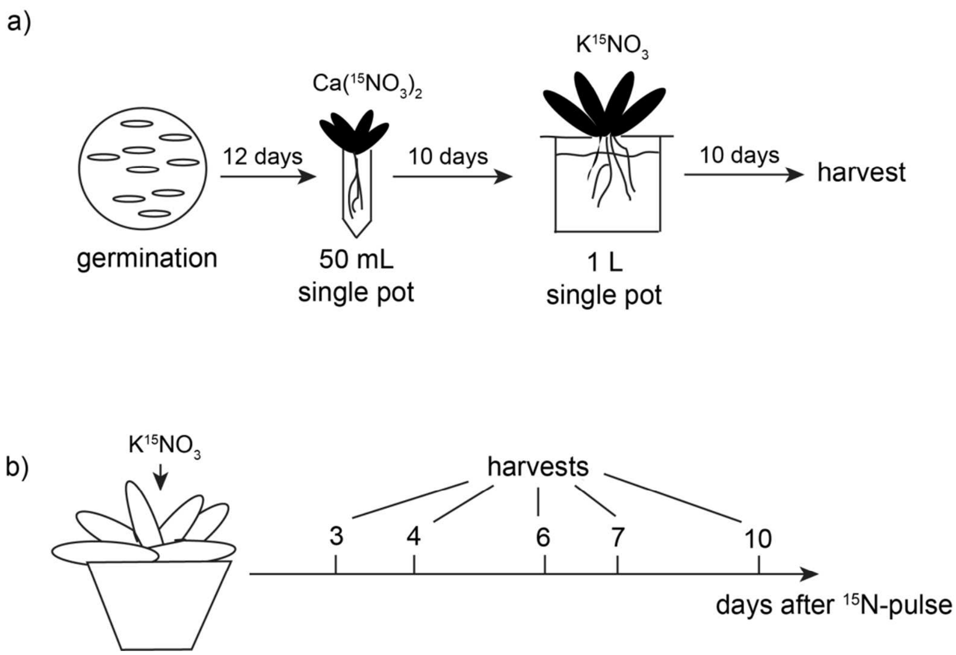

a)
K15NO3
Ca(15NO3)2
12 days
10 days
10 days
harvest
germination
50 mL
single pot
1 L
single pot
b)
K15NO3
harvests
3
4
6
7
10
days after 15N-pulse

Figure S2

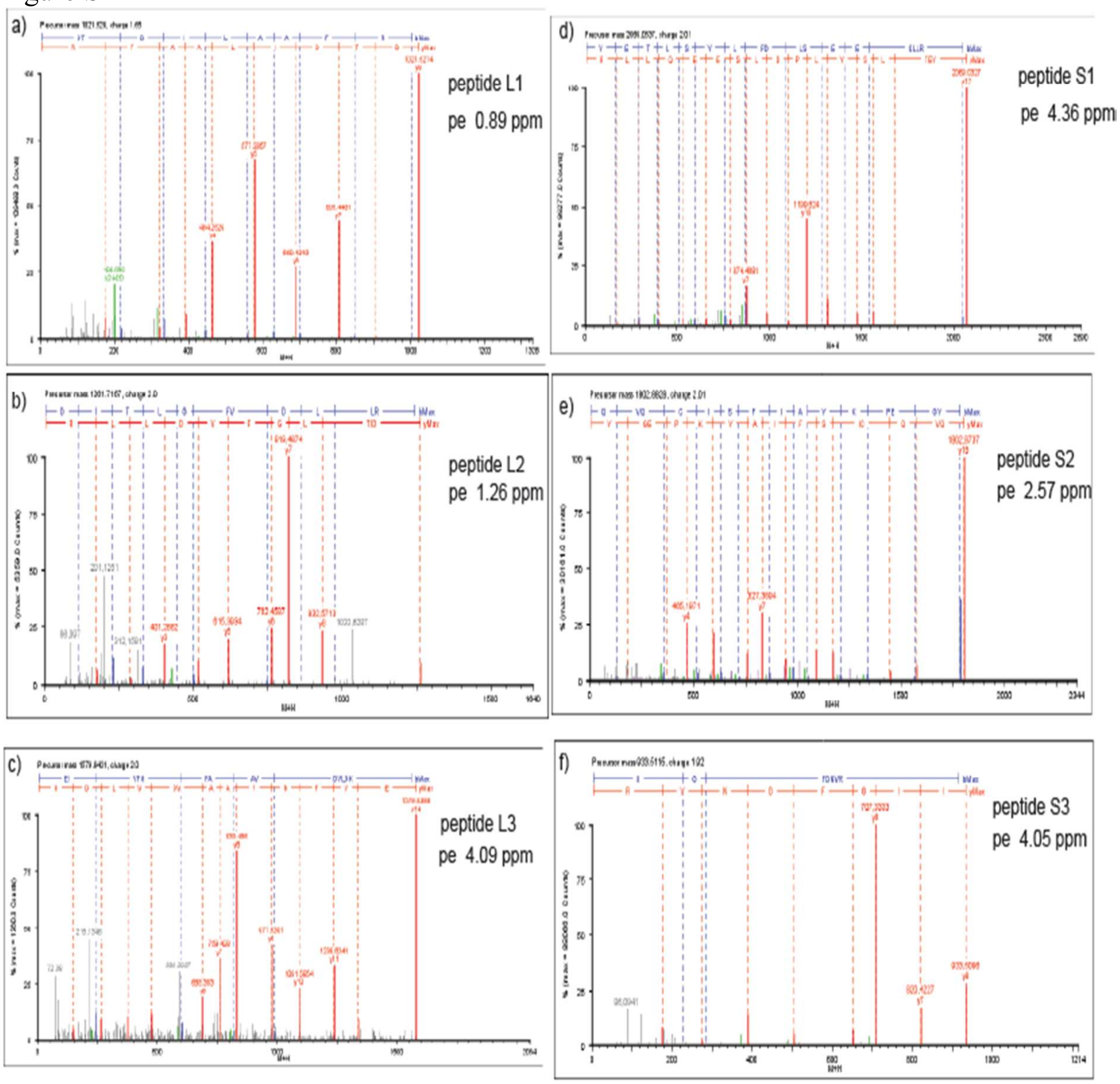

a)
peptide L1
pe 0.89 ppm
d)
peptide S1
pe 4.36 ppm
b)
peptide L2
pe 1.26 ppm
e)
peptide S2
pe 2.57 ppm
c)
peptide L3
pe 4.09 ppm
f)
peptide S3
pe 4.05 ppm

Figure S2 continued

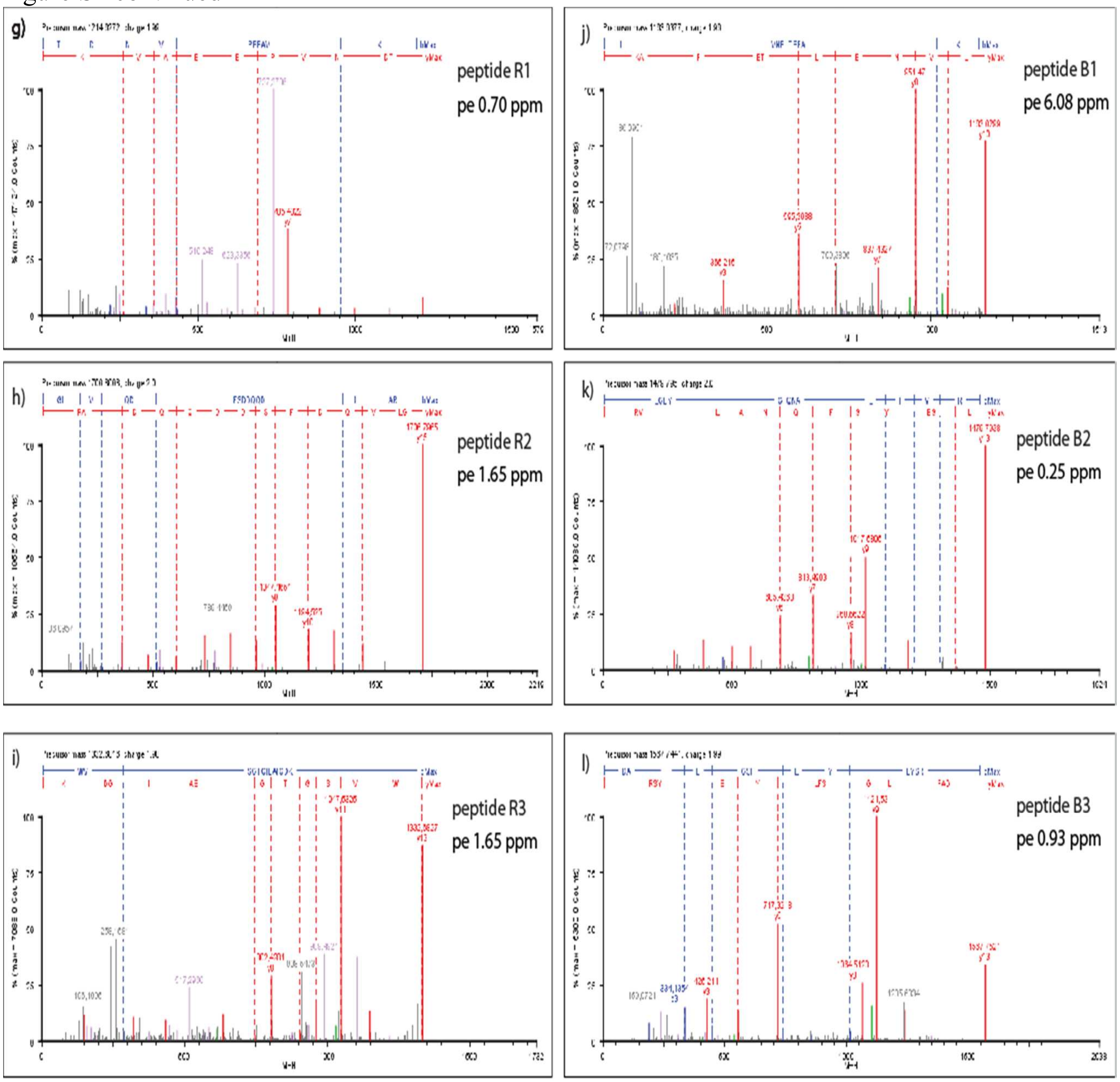
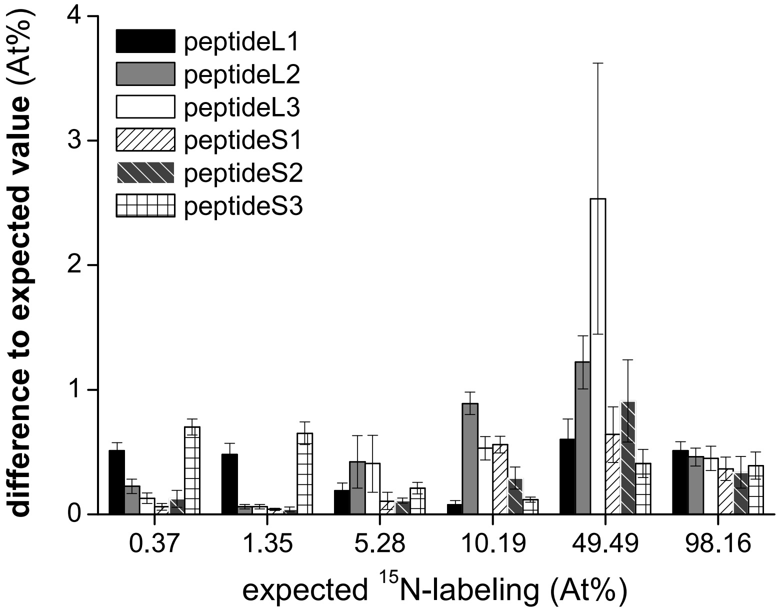
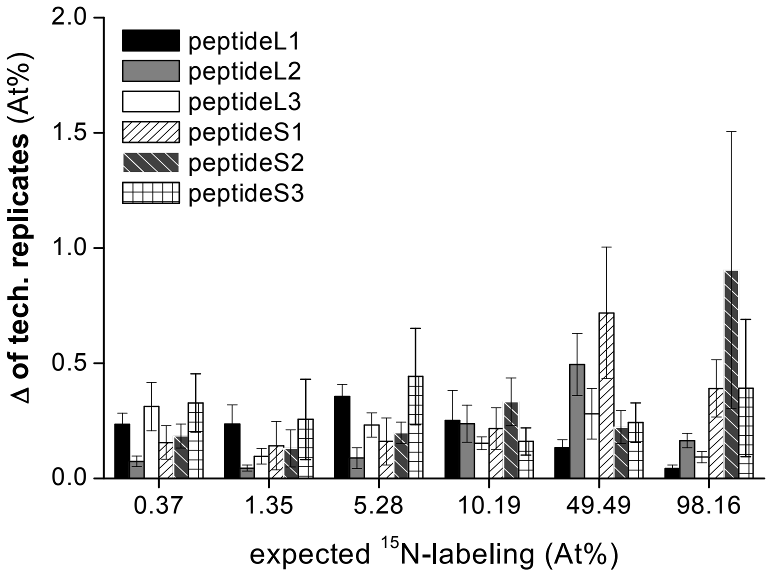


Figure S3



peptideL1
peptideL2
peptideL3
peptideS1
peptideS2
peptideS3
difference to expected value (At%)
0
1
2
3
4
0.37
1.35
5.28
10.19
49.49
98.16
expected 15N-labeling (At%)

Figure S4



peptideL1
peptideL2
peptideL3
peptideS1
peptideS2
peptideS3
Δ of tech. replicates (At%)
2.0
1.5
1.0
0.5
0.0
0.37
1.35
5.28
10.19
49.49
98.16
expected 15N-labeling (At%)

Figure S5

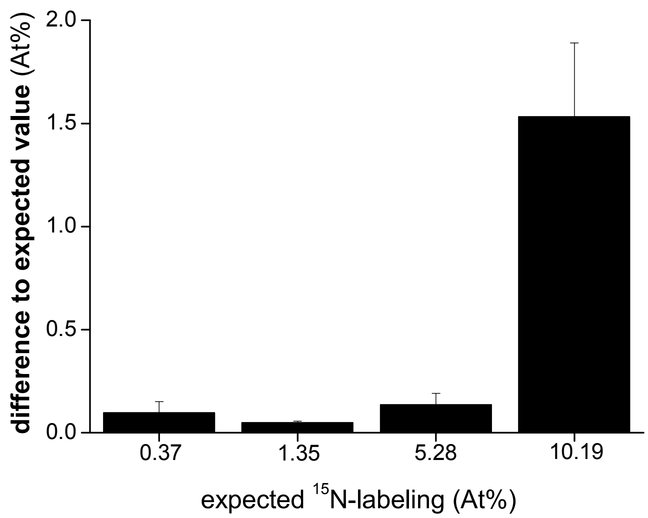

difference to expected value (At%)
2.0
1.5
1.0
0.5
0.0
0.37
1.35
5.28
10.19
expected 15N-labeling (At%)

Figure S6

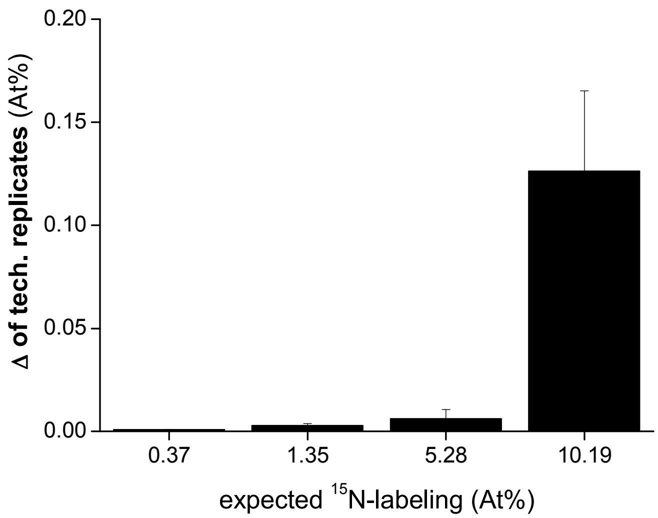
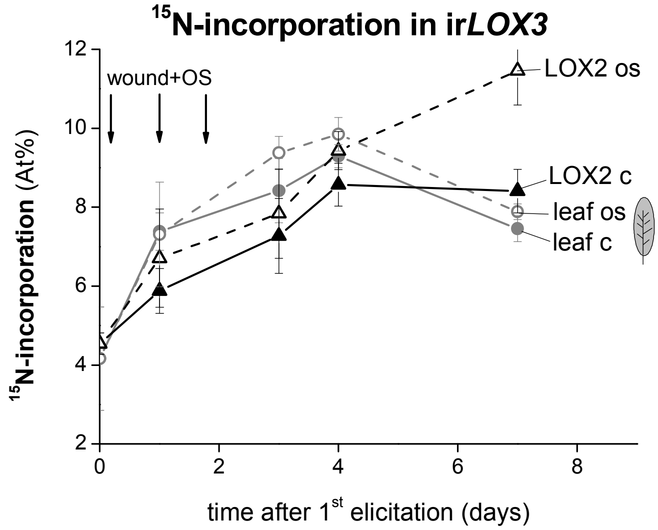

0.20
0.15
0.10
0.05
0.00
Δ of tech. replicates (At%)
0.37
1.35
5.28
10.19
expected 15N-labeling (At%)

Figure S7



^{15}N-incorporation in ir*LOX3*
wound+OS
LOX2 os
LOX2 c
leaf os
leaf c
^{15}N-incorporation (At%)
time after 1st elicitation (days)
12
10
8
6
4
2
0
2
4
6
8