Table S3. Quadrupole Moments of NPO and POM.

	NPO			POM			
Method	Q_{xx}	Q_{yy}	Q_{zz}	Q_{xx}	Q_{yy}	Q_{zz}	Q_{xz}
RHF/6-31G*	-48.21	-56.21	-84.55	-53.51	-62.39	-91.47	1.24
MP2(full)/6-31G*	-49.17	-56.18	-78.33	-54.77	-62.40	-84.68	1.11
Becke3LYP/6-31G*	-48.69	-55.19	-78.49	-53.94	-61.28	-84.37	1.22

(a) The symmetry axis of NPO coincided with the z-axis and the molecules lies in the xz-plane. POM lies in the xz plane as well and the z-axis coincides with the line connecting the NO nitrogen and the C atom to which the nitro group is attached.

(b) All values in Debye•Ångstrom (DÅ).

Table S4. Natural Population Analyses of NPO and POM.[a]

	NPO			POM		
Atom	RHF	MP2(full)	Becke3LYP	RHF	MP2(full)	Becke3LYP
N1(O)	0.14	0.10	0.10	0.15	0.10	0.10
C2	-0.01	-0.04	-0.04	-0.02	-0.03	-0.04
C2H	0.25	0.22	0.24	0.24	0.23	0.22
C3	-0.19	-0.25	-0.22	0.01	-0.04	-0.01
C4	0.02	0.04	0.04	0.01	0.05	0.04
C5	-0.19	-0.25	-0.22	-0.18	-0.25	-0.22
C5H	0.09	0.04	0.07	0.10	0.04	0.07
C6	-0.01	-0.04	-0.04	-0.02	-0.03	-0.04
C6H	0.25	0.22	0.24	0.24	0.23	0.22
O7	-0.59	-0.45	-0.46	-0.59	-0.46	-0.46
N8	0.66	0.47	0.51	0.66	0.47	0.51
O9	-0.46	-0.35	-0.38	-0.46	-0.35	-0.39
O10	-0.46	-0.35	-0.38	-0.46	-0.35	-0.39
Me/H11	0.28	0.29	0.29	0.10	0.11	0.09
Σ(NO)	-0.45	-0.35	-0.36	-0.44	-0.36	-0.36
$\Sigma(C_5H_3R)$	0.70	0.56	0.66	0.70	0.62	0.63
$\Sigma(NO_2)$	-0.26	-0.23	-0.25	-0.26	-0.23	-0.27

(a) All calculations employed the 6-31G* basis set.

Table S5. Molecular and Point-Charge-Model Derived Dipole Moments for NPO and POM.[a]

Method	α(°)	|μ|		$\|\mu\|_{NP}$[b]		$\|\mu\|_{MP}$[b]
NPO						
RHF/6-31G*		0.27		0.33		1.02
MP2(full)/6-31G*		0.95		1.14		1.30
Becke3LYP/6-31G*		1.25		1.49		1.65
POM						
RHF/6-31G*	93.5	0.80	78.8	0.88	60.6	1.37
MP2(full)/6-31G*	49.2	0.88	39.0	1.16	43.7	1.25
Becke3LYP/6-31G*	41.6	1.18	33.7	1.55	39.5	1.63

(a) Dipole moments in Debye.

(b) Values $|\mu|$ are the directly computed molecular dipole moments while the approximate dipole moments $|\mu|_{NP}$ and $|\mu|_{MP}$ are derived from point charge models as described in the text.