Supporting information for:

All-Solid-State Potentiometric Sensor Based on Graphene Oxide as Ion-to-Electron Transducer for Nitrate Detection in Water Samples

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KEYWORDS: *graphene oxide; ion-selective electrodes; potentiometry; nitrate; water*

Tables

Table S1. I_D/I_G ratios for CSPE/GO obtained from different surface zones.

Zone	I_D/I_G
1	0.98
2	0.95
3	0.99
4	1.05
5	1.02
6	0.99

Table S2. Repeatability assessment of the potentiometric response of five equally prepared CSPE/GO/ISM sensors (three subsequent calibrations on the same sensor unit).

Electrode no.	Slope (mV dec^{-1})	Intercept (mV)
1	–50.4	225.5
	–52.3	222.8
	–54.1	226.4
2	–51.8	206.6
	–52.4	207.1
	–57.4	200.2
3	–52.2	212.2
	–54.4	213.7
	–58.2	209.5
4	–53.4	213.6
	–52.8	218.3
	–52.9	217.7
5	–53.1	217.7
	–53.3	218.9
	–53.2	218.5
Average	**–53.5**	**215.2**
SD	**2.0**	**7.3**
CV	**3.8%**	**3.4%**

CV—Coefficient of variation; SD—Standard deviation.

Table S3. Reproducibility assessment of the potentiometric response of fifteen equally prepared CSPE/GO/ISM sensors (one calibration on each sensor unit).

Electrode no.	**Slope (mV dec^{-1})**	**Intercept (mV)**
1	–52.3	222.8
2	–52.4	207.1
3	–52.2	212.2
4	–50.6	213.6
5	–53.1	217.7
6	–49.8	223.9
7	–52.4	231.2
8	–52.0	211.3
9	–54.5	232.7
10	–53.3	228.2
11	–53.4	213.6
12	–53.1	217.7
13	–51.4	241.0
14	–53.4	242.9
15	–53.1	243.0
Average	**–52.3**	**223.9**
SD	**1.5**	**12.1**
CV	**2.8%**	**5.4%**

CV—Coefficient of variation; SD—Standard deviation.

Table S4. Comparison of the proposed all-solid-state NO_3^- ISE based on GO with published reports using different transducers.

Transducer	Recognition element	Slope (mV dec^{-1})	LOD (μM)	Response time (s)	Long-term drift (mV h^{-1})	Capacitance (μF)	Lifetime	Ref.
rGO	TDDMA-NO_3	–57.9	30	10	NR.	NR.	NR.	[35]
Hydrophobic LIG	TDDMA-NO_3	–58.2	6.0	12.5	0.33 ± 0.04	95.2 ± 8.2	5 weeks	[61]
f-MWCNTs	TDDMA-NO_3	–57.7	0.025	5	NR.	NR.	NR.	[26]
ErGO/AuNPs	PPy-NO_3	NR.	6.3	<15	NR.	5.1x10^{-3}	65 days	[56]
TTF-NO_3	NO_3 ionophore V	–59.4	0.63	NR.	NR.	60.3	NR.	[3]
TrGO	NO_3 ionophore VI	–60.0	4.0	<5	0.009	5.9x10^{-6}	2 weeks	[36]
MWCNTs	Nitron-NO_3	–55.1	0.028	<10	NR.	49.2	8 weeks	[34]
rGOA	TDDA-NO_3	–59.1	0.76	NR.	0.056	76.9	NR.	[37]
Graphene	TDDA-NO_3	–57.4	6.3	<3s	NR.	NR.	N.R.	[54]
GO	TDDA-NO_3	–53.5	1.9	10	0.32	32.6	3 months	This work

AuNPs: Gold nanoparticles; **ErGO**: Electrochemically reduced graphene oxide; **LIG**: Laser-induced graphene; **GO**: Graphene oxide; **MWCNTs**: Multiwalled carbon nanotubes; **NR.**: Not reported; **PPy**: Polypyrrole; **rGO**: Reduced graphene oxide; **rGOA**: Reduced graphene oxide aerogel; **TDDA-NO_3**: Tetradodecylammonium nitrate; **TDDMA-NO_3**: Tetradodecylmethylammonium nitrate; **TrGO**: Thiol-functionalized reduced graphene oxide; **TTF**: Tetrathiafulvalene.

Table S5. Potentiometric response characteristics of the proposed CSPE/GO/ISM sensors in 10^{-1} M phosphate buffer background (pH 5.0).

Parameter		
Slope (mV dec^{-1})		-51.5 ± 1.4
Intercept (mV)		228.2 ± 4.2
Coefficient of determination (R^2)		0.9979 ± 0.0023
Linear range of response (M)		$3.0 \times 10^{-6} - 10^{-2}$
Limit of detection (M)		2.1×10^{-6}
Response time (s)		10–20
Reproducibility slope (SD, mV)		
	Intra-electrode[1]	<1.4
	Inter-electrode[2]	2.4
Reproducibility intercept (SD, mV)		
	Intra-electrode[1]	<4.2
	Inter-electrode[2]	8.1

[1] Three consecutive calibrations performed on the same sensor; [2] Nine calibrations performed from three equally prepared sensors. SD—Standard deviation.

Figures

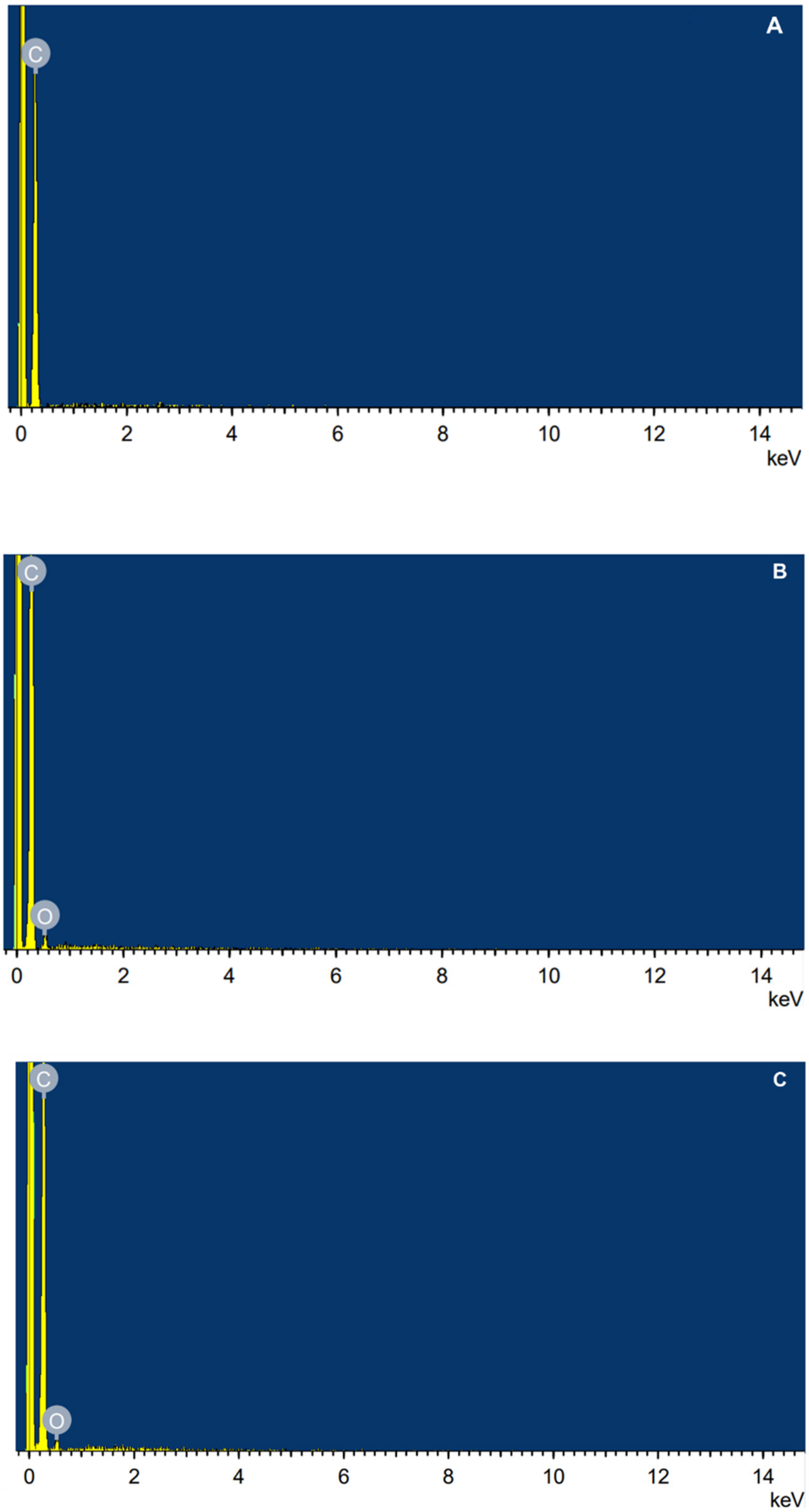

Figure S1. EDS analysis of the (**A**) CSPE bare electrode, (**B**) nanosheet and (**C**) particle in the CSPE/GO bare electrode.

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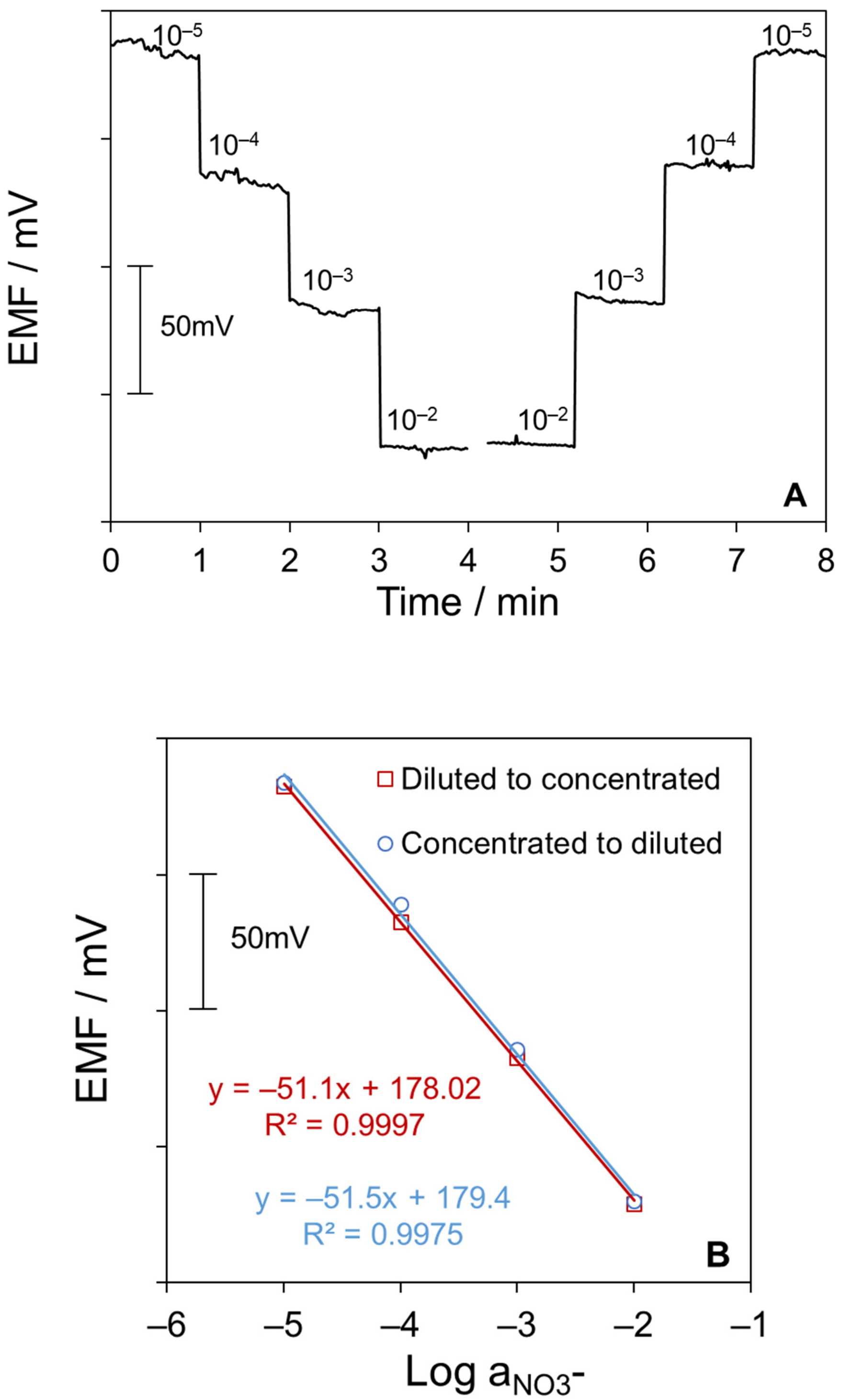

Figure S2. (**A**) Reversibility of CSPE/GO/ISM sensor by calibration from low to high analyte activities and vice-versa. (**B**) Corresponding calibrations graphs.

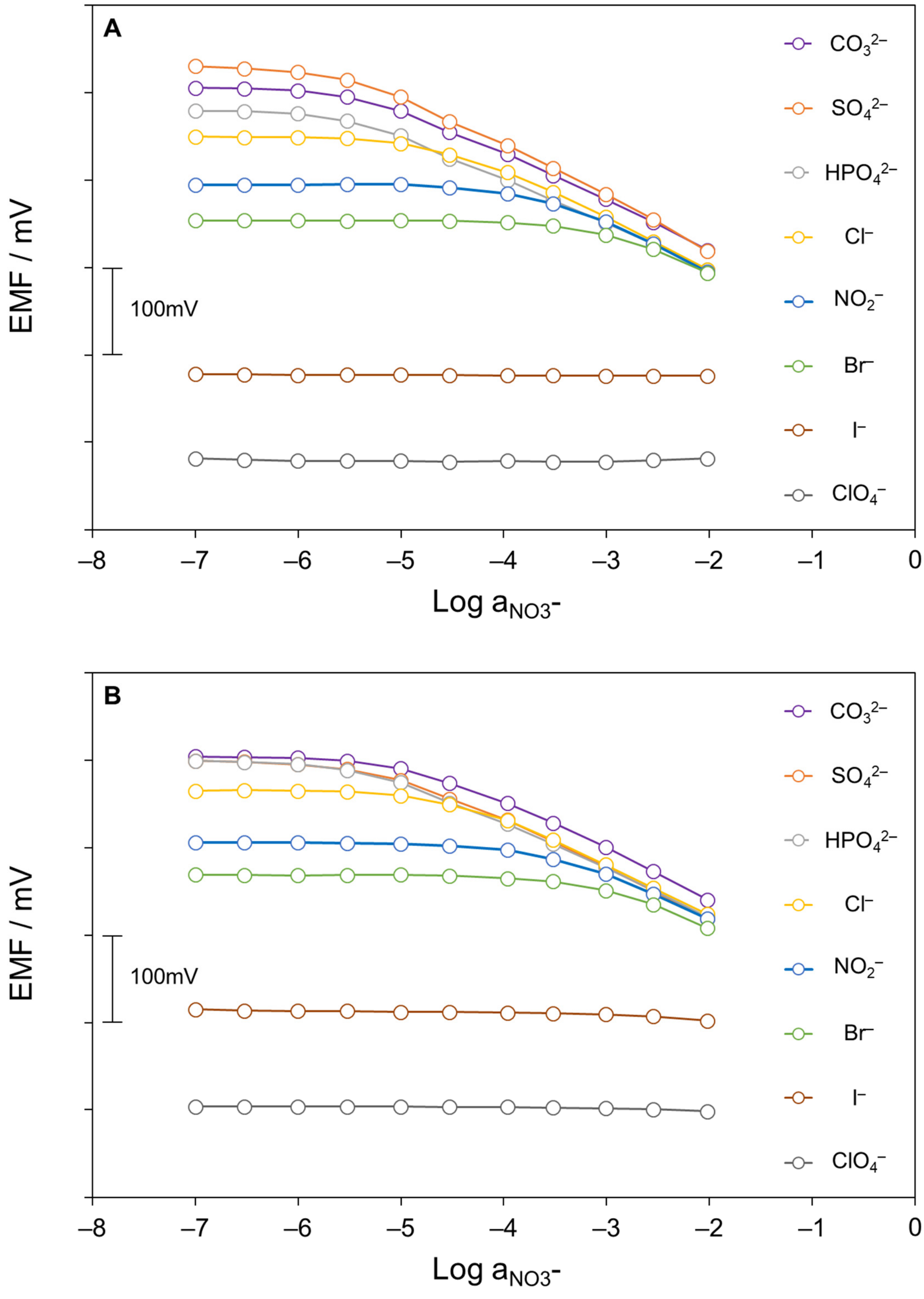

Figure S3. Potentiometric response of CSPE/ISM (**A**) and CSPE/GO/ISM (**B**) sensors towards the nitrate ion in the presence of different interfering anions at fixed concentration (10^{-2} M).

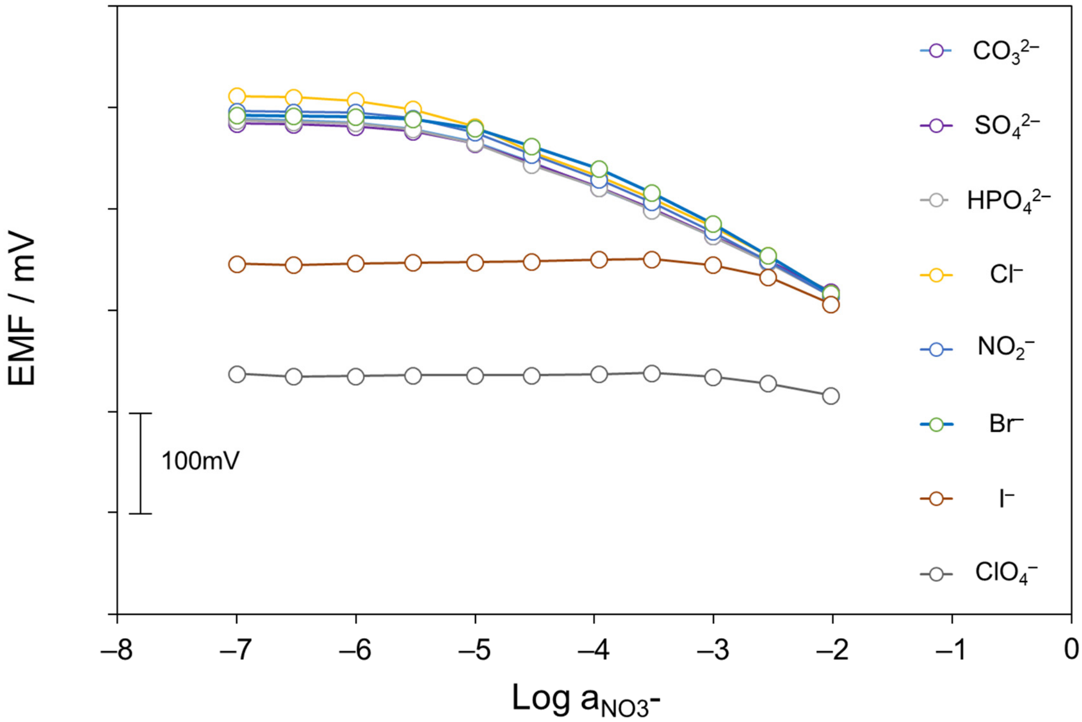

Figure S4. Potentiometric response of CSPE/GO/ISM sensors towards the nitrate ion in the presence of different interfering anions at fixed concentration (10^{-4} M).

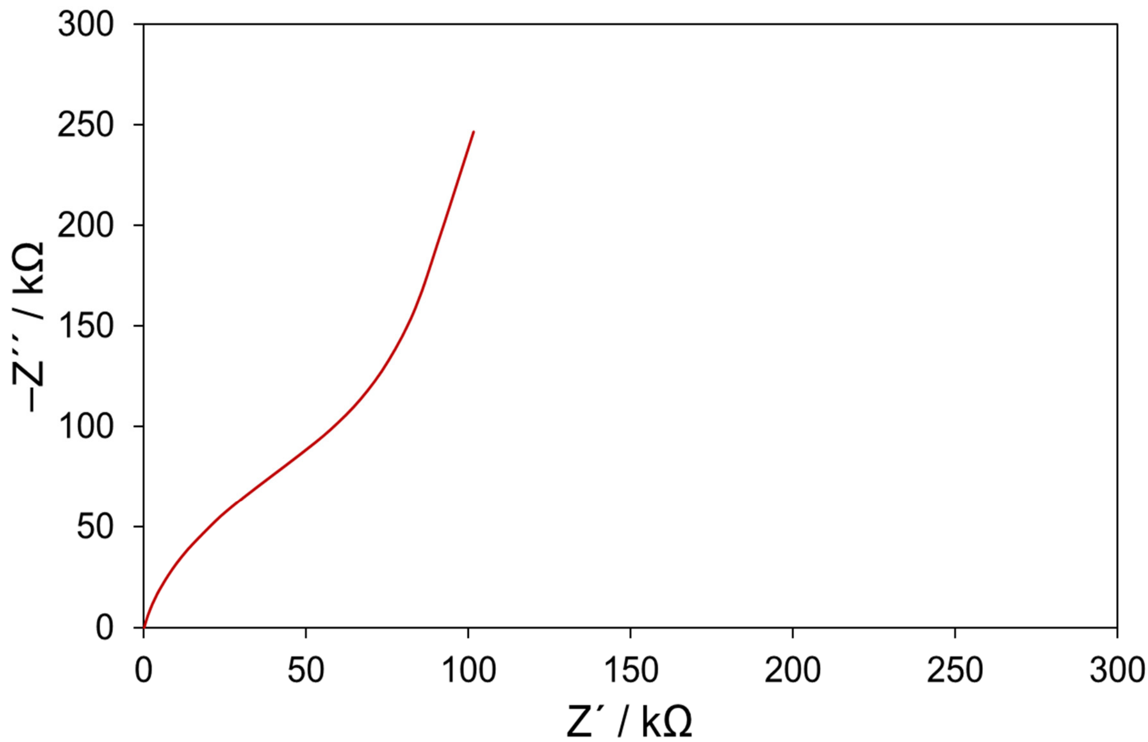

Figure S5. Nyquist plot of CSPE/GO bare electrode in 10^{-1} M $NaNO_3$ solution. Frequency range: 0.1 Hz to 100 kHz; E DC: OCP; ΔE AC: 10 mV.

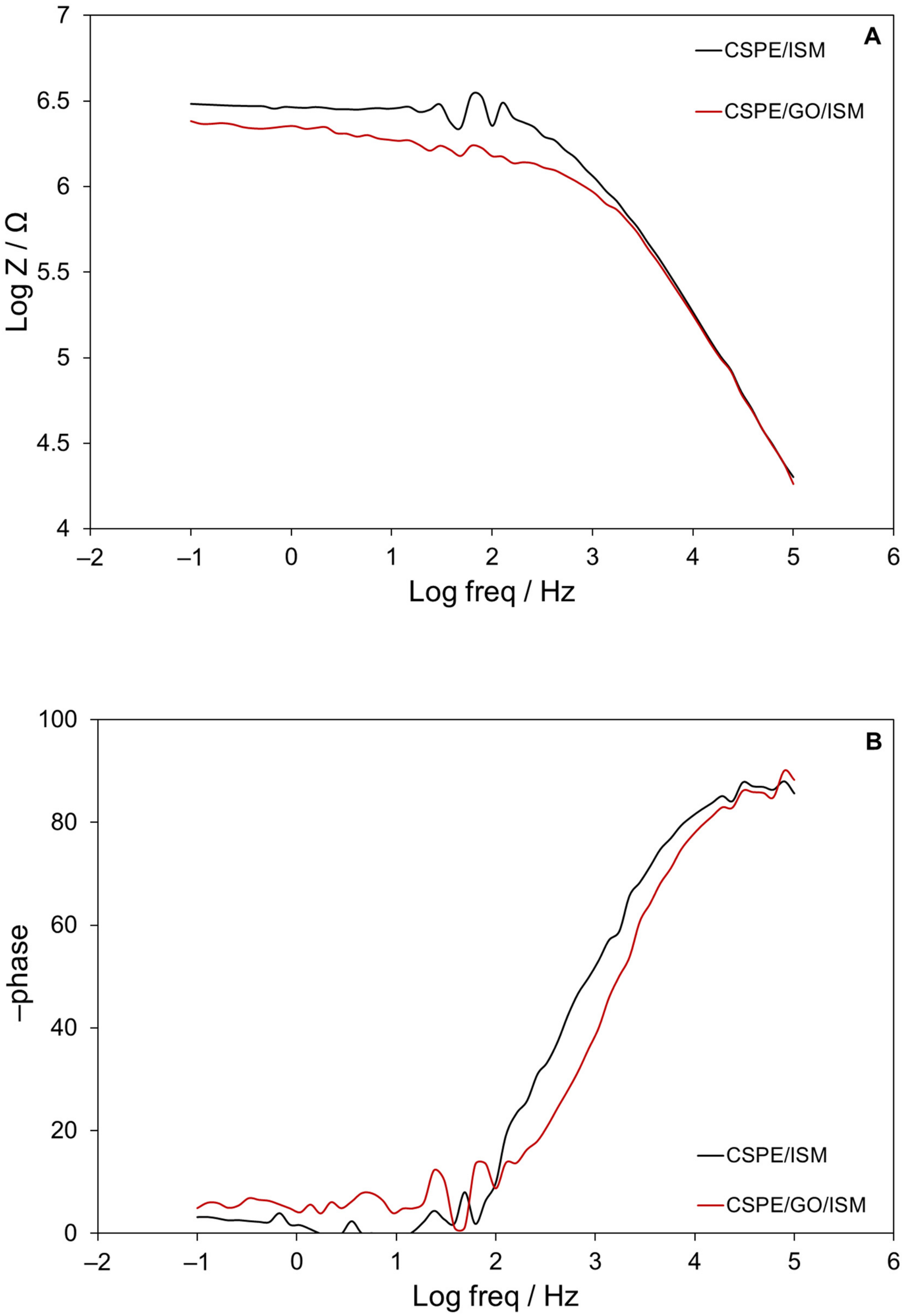

Figure S6. Electrochemical impedance bode plots for (**A**) impedance magnitude (Log Z) and (**B**) phase angle vs. Log frequency (f) in 10^{-1} M $NaNO_3$ solution. Frequency range: 0.1 Hz to 100 kHz; E DC: OCP; ΔE AC: 10 mV.

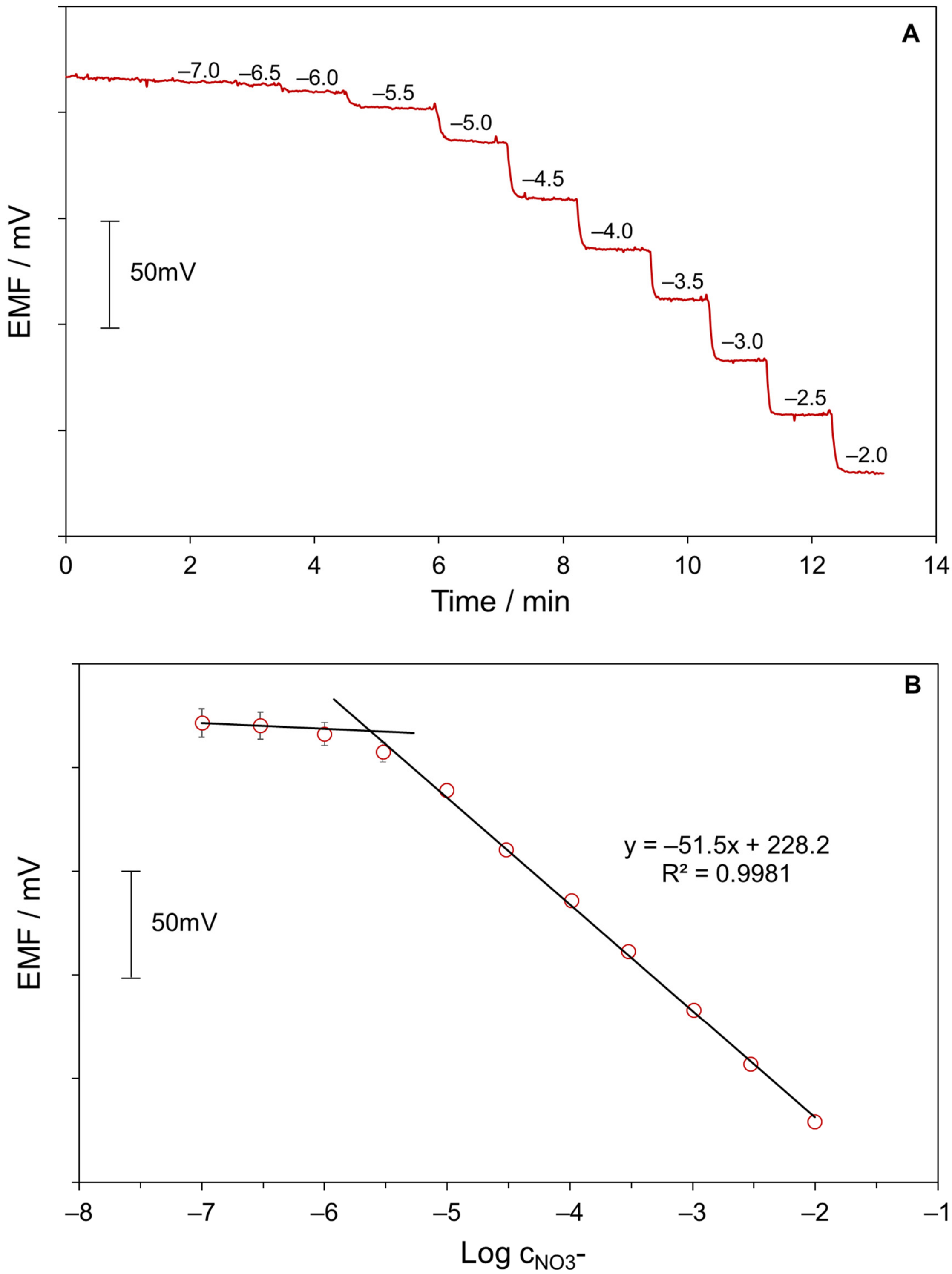

Figure S7. (**A**) Dynamic response of one CSPE/GO/ISM sensor in steady-state mode at increasing NO_3^- concentrations and using the commercial Ag/AgCl reference electrode (logarithmic concentrations are indicated above each trace). (**B**) Corresponding calibration graph whose error bars refer to the average of three successive calibrations. Background: phosphate buffer 10^{-1} M at pH 5.0 ($I = 10^{-1}$ M).

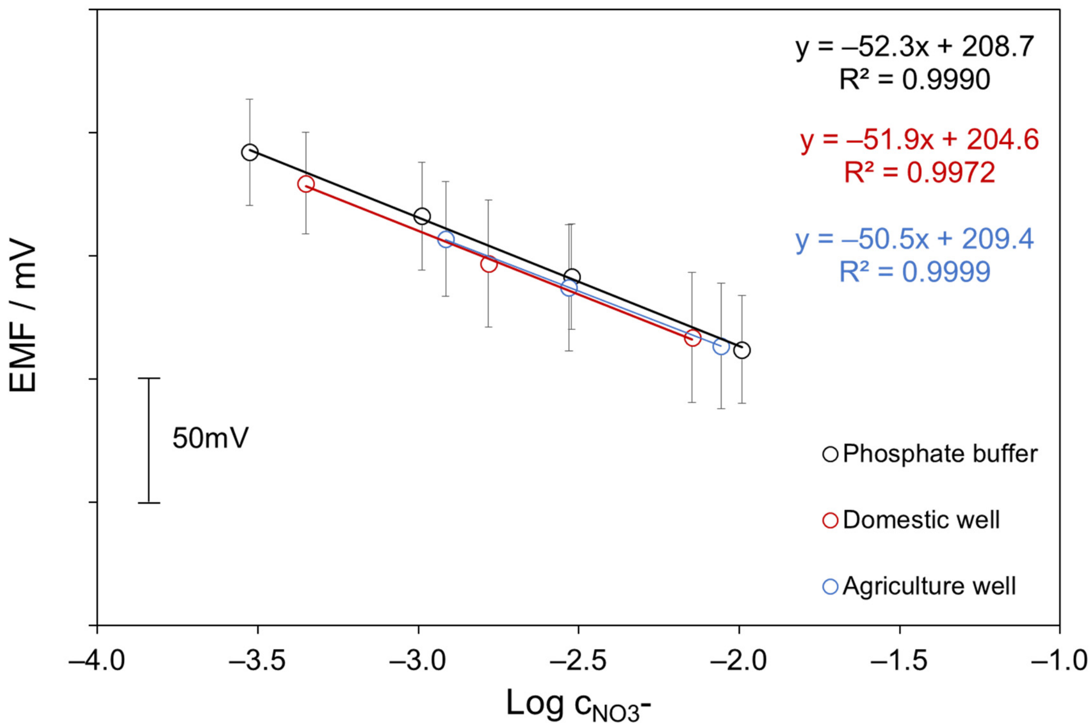

Figure S8. Dynamic calibration curves of CSPE/GO/ISM sensors towards the nitrate ion in different backgrounds (phosphate buffer and well water samples).

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