Supporting Information:

NMRlipids IV: Headgroup & glycerol backbone structures, and cation binding in bilayers with PE and PG lipids

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S1 Comparison of headgroup order parameters from different force fields to experiments

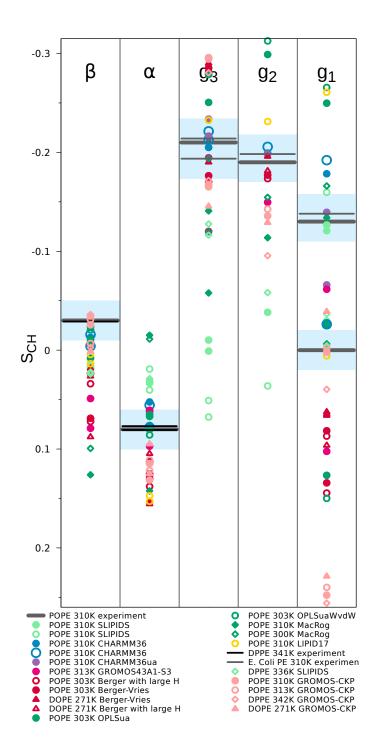


Figure S1: The headgroup and glycerol backbone order parameters of PE lipids from experiments (POPE and signs this work, DPPE from Ref. ? and E.coliPE from Ref. ?) and simulations with different force fields.

1. This should be clarified as in NMRlipidsI and error bars should be added. Probably larger error bars for united atom models based on the report by Fuchs et al.

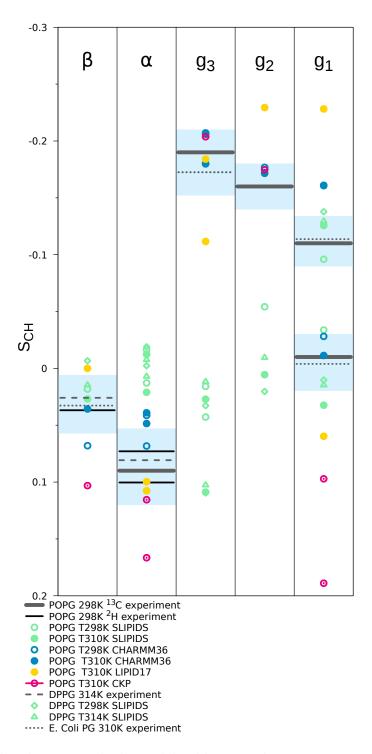


Figure S2: The headgroup and glycerol backbone order parameters of PG lipids from experiments (POPG and signs from this work and from Ref. ? , DPPG with 100mM NaCl from Ref. ? ,and E.Coli PG results from Ref. ?). and simulations with different force fields.

S2 Simulated systems

S2.1 CHARMM36

Table S1: List of MD simulations with PE lipids.

					ı				ı					1	ı		ı		ı				ı	
gffles	٠.	٠.	٠.	٠,	٠.	٠.	٠.	٠.	٠.	٠.	٠,	٠.	٠,	٠.	٠	٠.	٠.	٠.	٠.	٠.	٠.	٠.	٠.	٠.
$f_{\rm tanal}$ (ns)	400	100	100	2×100	100	2×100	100	100	2×400	2×400	100	100	2×400	2×100	2×100	2×100	350	300	2×100	2×100	2×100	2×100	100	100
$^{e}\mathrm{t_{sim}(ns)}$	200	500	500	2×200	200	2×200	200	200	2×500	2×500	200	200	2×500	2×200	2×200	2×200	200	200	2×200	2×200	2×200	2×300	200	200
$^{d}\mathrm{T}\left(\mathrm{K}\right)$	310	310	310	310	336	310	310	310	342	313	310	310	271	313	303	303	310	300	303	303	271	271	310	310
$^c{ m N}_{ m c}$	0	0	20	0	0	0	0	50	0	0	0	20	0	0	0	0	0	0	0	0	0	0	20	20
$^b\mathrm{N}_\mathrm{w}$	2760	25000	25000	15254	9386	~	25000	25000	3655	3552	25000	25000	4789	3552	3328	3328	2760	5120	3552	3552	4789	4789	25000	25000
$^a\mathrm{N}_1$	144	200	200	336	288	336	500	500	128	128	200	200	128	128	128	128	144	128	128	128	128	128	200	200
NaCl (M)	0	0	0.11	0	0	0	0	0.11	0	0	0	0.11	0	0	0	0	0	0	0	0	0	0	0	0.11
force field for lipids / ions	CHARMM36?	CHARMM36?	CHARMM36?	$ m CHARMM36ua^{2}$	Slipids?	Slipids?	Slipids?	Slipids / Åqvist??	GROMOS-CKP?	GROMOS-CKP?	GROMOS-CKP?	GROMOS-CKP?	GROMOS-CKP?	GROMOS 43A1-S3?	OPLS-UA vdW on H?	OPLS-UA?	$OPLS-MacRog^{?}$	$\mathrm{OPLS ext{-}MacRog}^{?}$	Berger-Vries?	$\operatorname{Berger-largeH}^?$	Berger-Vries?	${ m Berger-largeH}^{?}$	LIPID17?	$LIPID17^{?}$
lipid/counter-ions	POPE	POPE	POPE	POPE	DPPE	POPE	POPE	POPE	DPPE	POPE	POPE	POPE	DOPE	POPE	POPE	POPE	POPE	POPE	POPE	POPE	DOPE	DOPE	POPE	POPE

 $[^]a\mathrm{Number}$ of lipid molecules with largest mole fraction

8.LIPID17 simulations with correct dihedrals still coming

 $[^]b\mathrm{Number}$ of water molecules $^c\mathrm{Number}$ of additional cations

 $[^]d {\rm Simulation\ temperature} \\ ^e {\rm Total\ simulation\ time}$ f Time used for analysis

 $[^]g$ Reference for simulation files

^{7.} Citations for Berger-* simulations? 5. Citation for GROMOS 43A1-S3? 6. Citation for OPLS-UA models? 2. Citation for CHARMM36 PE? 3. Which ion model is used in? ? 4. Citation for GROMOS-CKP?

Table S2: List of MD simulations with PG lipids.

				1					1		1	
gfiles	٠.	٠.	٠.	٠	٠.	¢.	٠.	٠.	٠.	٠.	٠.	٠.
$f_{\rm t_{anal}}$ (ns)	100	100	100	100	100	. 100	100	100	100	100?	100°	100
•												
$^{e}\mathrm{t_{sim}(ns)}$	100	200	500	250	200	400	500	500	200	200	200	200
^{d}T (K)		310	310	298	314	298	310	310	310	310	310	310
c N $_c$	0	49	0	0	0	0	0	49	0	49	0	49
$^b\mathrm{N}_\mathrm{w}$	4110	25000	25000	10664	11232	11232	25000	25000	25000	25000	25000	25000
$^a\mathrm{N}_1$	118	500	200	288	288	288	200	200	200	200	200	500
NaCl (M)	0	0.11	0	0	0	0	0	0.11		0.11		0.11
force field for lipids / ions	CHARMM36? 9.	CHARMM36?	CHARMM36?	Slipids / Åqvist??	Slipids / Åqvist??	Slipids / Åqvist??	Slipids / Åqvist??	POPG Slipids / Åqvist??	$LIPID17 / Dang^{?}$	LIPID17?	GROMOS-CKP?	GROMOS-CKP?
lipid/counter-ions	$POPG/K^+$	POPG	POPG	POPG/Na+	$DPPG/Na^{+}$	$DPPG/Na^{+}$	POPG	POPG	POPG	POPG	POPG	POPG

 $[^]a\mathrm{Number}$ of lipid molecules with largest mole fraction

10. Citations and ion model for CHARMM36?

12. Citation and ion model for GROMOS-CKP?

 $[^]b\mathrm{Number}$ of water molecules $^c\mathrm{Number}$ of additional cations

 $^{^{}d} {\rm Simulation\ temperature} \\ ^{e} {\rm Total\ simulation\ time}$

 $f{\rm Time~used~for~analysis}$ $g{\rm Reference~for~simulation~fles}$

^{11.}Lipid17 simulation with correct dihedral potentials still coming.

Table S3: List of MD simulations with PE and PG lipids mixed with PC.

POPC CHARMM36? 0 500 2500 0 310 500 100 7 POPC:POPG (1:1) CHARMM36? 0 150.150 3150 0 298 500 400 7 POPC:POPG (1:1) CHARMM36? 0 0.1 150.150 3150 0 298 500 400 7 POPC:POPG (1:1) CHARMM36? 0 0.1 150.150 2976 578 298 500 400 7 POPC:POPG (4:1) CHARMM36? 0 0.1 350.88 26280 7 298 500 400 7 POPC:POPG (4:1) CHARMM36? 0 0.1 350.88 26280 47 298 500 400 7 POPC:POPG (4:1) CHARMM36? 0 0.1 350.88 26280 47 300 300 250 400 7 POPC CHARMM36? 0 0.1 236.88 26280 47 30 300		ione mend not inbine / ione	NaCI (M)	CaC12 (1VI)		M		(V)	\log_{11S}	$^{\prime} t_{\rm anal} ({ m ns})$	$^{\circ}$ IIIeS
POPG (7:3) CHARMM36? 0 350 250 0 310 500 POPG (1:1) CHARMM36? 0 150:150 31500 0 298 500 POPG (1:1) CHARMM36? 0 0.1 150:150 3176 57 298 400 POPG (1:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36? 0 0.1 350:88 24927 451 298 500 POPG (4:1) CHARMM36? 0 0.1 256 8704 0 300 POPG (4:1) CHARMM36? 0 0 128 8704 0 300 POPG (4:1) CHARMM36? 0 0 128 8704 0 300 POPG (1:1) CHARMM36? 0 0 128 5120 0	POPC	CHARMM36?	0	0	200	25000	0	310	200	100	٠.
POPG (1:1) CHARMM367 0 150:150 3150 0 298 500 POPG (1:1) CHARMM367 0 0.1 150:150 31329 57 298 400 POPG (1:1) CHARMM367 0 0.1 150:150 2766 578 298 500 POPG (4:1) CHARMM367 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM367 0 0.1 350:88 24927 451 298 500 POPG (4:1) CHARMM367 0 0 1.0 350:88 24927 451 298 500 POPE (1:1) CHARMM367 0 0 128 8704 0 300 300 POPE (1:1) CHARMM367 0 0 128 8704 0 300 300 POPE (1:1) CHARMM367 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36	POPC:POPG (7:3)	${ m CHARMM36}^{?}$	0	0	350	25000	0	310	200	100	٠,
POPG (1:1) CHARMM36? 0 0.1 150:150 3129 57 298 400 POPG (1:1) CHARMM36? 0 1.08 150:150 29766 578 298 500 POPG (4:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPE (1:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPE (1:1) CHARMM36? 0 0.1 128 8704 0 300 300 POPE (1:1) OPLS-MacRog? 0 0.1 128 8704 0 300 500 POPE (1:1) OPLS-MacRog? 0 0.1 128 5120 0 238 500 POPE (1:1)	POPC:POPG (1:1)	${ m CHARMM36}^{?}$	0	0	150:150	31500	0	298	200	400	٠
POPG (1:1) CHARMM36² 0 1.08 150:150 29766 578 298 500 POPG (4:1) CHARMM36² 0 0.1 350:88 26280 0 298 500 POPG (4:1) CHARMM36² 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36² 0 0.1 350:88 24927 451 298 500 POPE (1:1) CHARMM36² 0 0.1 256 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 5120 0 298 500 POPE (1:1) Silpid / Dang²²²²²	POPC:POPG (1:1)	CHARMM36?	0	0.1	150:150	31329	22	298	400	300	٠.
POPG (4:1) CHARMM36² 0 350:88 26280 0 298 500 POPG (4:1) CHARMM36² 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36² 0 0.1 350:88 24927 451 298 500 POPG (4:1) CHARMM36² 0 0 256 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36² 0 0 128 5120 0 300 500 POPE (1:1) Slipid² 0 0 128 5120 0 300 500 POPE (1:1) Slipid² 0 0 128 5120 0 30 500 POPG (7:3) GROMOS-CKP / ????? 0 5	POPC:POPG (1:1)	${ m CHARMM36}^{?}$	0	1.08	150:150	29766	578	298	200	400	٠
POPG (4:1) CHARMM36? 0 0.1 350:88 26280 47 298 500 POPG (4:1) CHARMM36? 0 1.0 350:88 24927 451 298 500 POPE (1:1) CHARMM36? 0 0 256 8704 0 300 300 POPE (1:1) CHARMM36? 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36? 0 0 128 5120 0 300 500 POPE (1:1) CHARMM36? 0 0 128 5120 0 300 500 POPE (1:1) Slipid? 0 128 5120 0 298 500 POPE (1:1) Slipid? 0 0 500 2500 0 310 500 POPG (7:3) GROMOS-CKP / ???? 0 350:150 2500 0 310 500 POPG (7:3) Slipid / Åqvist.?. 0 350:150 <td>POPC:POPG (4:1)</td> <td>${ m CHARMM36}^{?}$</td> <td>0</td> <td>0</td> <td>350.88</td> <td>26280</td> <td>0</td> <td>298</td> <td>200</td> <td>400</td> <td>٠,</td>	POPC:POPG (4:1)	${ m CHARMM36}^{?}$	0	0	350.88	26280	0	298	200	400	٠,
POPG (4:1) CHARMM36? 0 1.0 350:88 24927 451 298 500 POPE (1:1) CHARMM36? 0 1.0 256 8704 0 300 300 POPE (1:1) CHARMM36? 0 0 128 8704 0 300 300 POPE (1:1) CHARMM36? 0 0 128 5120 0 300 500 POPE (1:1) OPLS-MacRog? 0 0 128 5120 0 300 500 POPE (1:1) Slipid? 0 0 128 5120 0 300 500 POPE (1:1) Slipid? 0 0 128 5120 0 298 500 POPG (7:3) GROMOS-CKP / ???? 0 0 350.150 2500 0 310 500 POPG (7:3) Slipid / Dang???? 0 0 350.150 2500 0 310 500 POPG (1:1) Slipid / Dang????<	POPC:POPG (4:1)	${ m CHARMM36}^{?}$	0	0.1	350:88	26280	47	298	200	400	٠,
CHARMM36° O	POPC:POPG (4:1)	${ m CHARMM36}^{?}$	0	1.0	350:88	24927	451	298	200	400	٠.
POPE (1:1) CHARMM367 0 0 128 8704 0 300 300	POPC	CHARMM36?	0	0	256	8704	0	300	300	250	٠.
OPLS-MacRog? 0 128 5120 0 500 POPE (1:1) OPLS-MacRog? 0 0 128 5120 0 500 POPE (1:1) Slipid² 0 0 512 23943 0 500 POPE (1:1) Slipid² 0 0 512 23943 0 298 170 POPE (1:1) Slipid² 0 0 0 128 5120 0 298 500 POPG (7:3) GROMOS-CKP / ??? ? 0 0 350:150 25000 0 310 500 POPG (7:3) Slipid / Dang² ? ? ? 0 0 350:150 25000 0 310 500 POPG (1:1) Slipid / Dang² ? ? ? 0 0 128:128 12800 0 298 500 POPG (1:1) Slipid / Dang² ? ? ? 0 0 128:128 12800 46 298 1500 POPG (1:1) Slipid / Dang² ? ? ? 0 0	POPC:POPE (1:1)	${ m CHARMM36}^{?}$	0	0	128	8704	0	300	300	250	٠٠
POPE (1:1) OPLS-MacRog² 0 128 5120 0 300 500 POPE (1:1) Slipid² 0 0 512 23943 0 298 170 POPE (1:1) Slipid² 0 0 128 5120 0 298 500 POPE (1:1) Slipid² 0 0 500 2500 0 310 500 POPG (7:3) GROMOS-CKP / ??? ? 0 0 350:150 25000 0 310 500 POPG (7:3) Slipid / Åqvist² ? 0 0 350:150 25000 0 310 500 POPG (1:1) Slipid / Dang² ? ? ? 0 0 128:128 12800 0 298 500 POPG (1:1) Slipid / Dang² ? ? ? 0 0 128:128 12800 46 298 1500 POPG (1:1) Slipid / Dang² ? ? ? 0 0 0 128:128 12800 46 298 1500 P	POPC	OPLS-MacRog?	0	0	128	5120	0	300	200	300	ç.
Slipid Slipid O	POPC:POPE (1:1)	$\mathrm{OPLS ext{-}MacRog}^{?}$	0	0	128	5120	0	300	200	300	٠.
CROMOS-CKP ??? 0	POPC	Slipid?	0	0	512	23943	0	298	170	100	٠.
GROMOS-CKP / ?? ? ° ° ° 500 25000 0 310 500 FOPG (7:3) GROMOS-CKP / ?? ? ° ° ° ° ° 550:150 25000 0 310 500 Slipid / Dang ? ? ? ° ° ° ° ° 550:150 25000 0 310 500 FOPG (1:1) Slipid / Dang ? ? ? ° ° ° ° ° ° ° ° ° ° ° ° ° ° ° °	POPC:POPE (1:1)		0	0	128	5120	0	298	200	300	٠.
POPG (7:3) GROMOS-CKP / ?? ? 0 0 350:150 25000 0 310 500 POPG (7:3) Slipid / Åqvist ? ? 0 0 350:150 25000 0 310 500 POPG (1:1) Slipid / Dang ? ? ? 0 0 128:128 12800 0 298 500 POPG (1:1) Slipid / Dang ? ? ? 0 0.1 128:128 12800 46 298 1500 POPG (1:1) Slipid / Dang ? ? ? 0 0.5 128:128 12800 46 298 1500 POPG (1:1) Slipid / Dang ? ? ? 0 0.5 128:128 12800 230 298 1500 POPG (1:1) Slipid / Dang ? ? ? 0 1.0 128:128 12800 298 1500	POPC	_	0	0	200	25000	0	310	200	100	٠.
Slipid ² 0 0 500 25000 0 310 500 POPG (7:3) Slipid / Åqvist ² ? 0 0 0 128:128 12800 0 298 500 POPG (1:1) Slipid / Dang ² ? ? 0 0 0.1 128:128 12800 23 298 500 POPG (1:1) Slipid / Dang ² ? ? 0 0 0.2 128:128 12800 46 298 1500 POPG (1:1) Slipid / Dang ² ? ? 0 0 0.5 128:128 12800 415 298 1500 POPG (1:1) Slipid / Dang ² ? ? 0 0 0.5 128:128 12800 230 298 1500 POPG (1:1) Slipid / Dang ² ? ? 0 0 1.0 128:128 12800 230 298 1500	POPC:POPG (7:3)	_	0	0	350:150	25000	0	310	200	100	٠
Slipid / Åqvist?? 0 0 350:150 25000 0 310 500 500 Slipid / Dang???? 0 0 128:128 12800 0 298 500 500 Slipid / Dang???? 0 0 0.1 128:128 12800 23 298 500 500 Slipid / Dang???? 0 0 0.2 128:128 12800 46 298 1500 Slipid / Dang???? 0 0 0.5 128:128 12800 115 298 1500 Slipid / Dang???? 0 1.0 128:128 12800 230 298 1500 500 Slipid / Dang????	POPC	Slipid?	0	0	200	25000	0	310	200	100	٠.
(1:1) Slipid / Dang???? 0 0 128:128 12800 0 298 500 (1:1) Slipid / Dang???? 0 0.1 128:128 12800 23 298 500 (1:1) Slipid / Dang???? 0 0.2 128:128 12800 46 298 1500 (1:1) Slipid / Dang???? 0 0.5 128:128 12800 115 298 1500 (1:1) Slipid / Dang???? 0 1.0 128:128 12800 230 298 1500 (1:1) Slipid / Dang????	POPC:POPG (7:3)	Slipid / Åqvist??	0	0	350:150	25000	0	310	200	100	٠.
(1:1) Slipid / Dang???? 0 0.1 128:128 12800 23 298 500 (1:1) Slipid / Dang???? 0 0.2 128:128 12800 46 298 1500 (1:1) Slipid / Dang???? 0 0.5 128:128 12800 115 298 1500 (1:1) Slipid / Dang???? 0 1.0 128:128 12800 230 298 1500 (1:1)	POPC:POPG (1:1)		0	0	128:128	12800	0	298	200	400	٠.
(1:1) Slipid / Dang [?] ? ? ° ° ° 0.2 128:128 12800 46 298 1500 (1:1) Slipid / Dang [?] ? ? ° ° ° 0.5 128:128 12800 115 298 1500 (1:1) Slipid / Dang [?] ? ? ° ° ° ° ° ° ° ° ° ° ° ° ° ° ° ° °	POPC:POPG (1:1)		0	0.1	128:128	12800	23	298	200	400	٠.
(1:1) Slipid / Dang? ? ? ? 0 0.5 128:128 12800 115 298 1500 (1:1) Slipid / Dang? ? ? ? 0 1.0 128:128 12800 230 298 1500	POPC:POPG (1:1)	_	0	0.2	128:128	12800	46	298	1500	200	٠.
(1:1) Slipid / $Dang^{?}$? ? 0 1.0 128:128 12800 230 298 1500		\	0	0.5	128:128	12800	115	298	1500	200	٠.
	POPC:POPG (1:1)	_	0	1.0	128:128	12800	230	298	1500	200	٠.

 $[^]a\mathrm{Number}$ of lipid molecules with largest mole fraction

13.Citation and ion model for GROMOS-CKP?

14. Citation and description for "Berger" model?

15.Lipid17 POPC and POPC:POPG mixtures (https://doi.org/10.5281/zenodo.3241242 and https://doi.org/10.5281/zenodo.3237656) should be added

16.Upcoming Lipid17ecc with POPC:POPS (4:1) mixture simulations to be added.

after simulated with corrected dihedrals.

 $^{{}^}b\mathrm{Number}$ of water molecules

 $^{^{}c}$ Number of additional cations

 $[^]d$ Simulation temperature

 $[^]e$ Total simulation time

 $[^]f\Gamma$ ime used for analysis

 $[^]g$ Reference for simulation files

Table S4: List of MD simulations with PE and PG lipids mixed with PC.

ns) gfiles	٠.	٥.	٥٠	٥.	٠.	٥.	٠.	٥٠	ر.	٠.	٠.	¢•	٠.	٠.
$f_{\rm tanal} $ (ns)	350	250	200	200	198	200	333	300	400	200	200	200	200	200
$^{e}\mathrm{t}_{\mathrm{sim}}\mathrm{(ns)}$	400	400	1200	320	718	720	347.8	400	009	300	300	300	300	300
$^{d}\mathrm{T}$ (K)	298	298	298	298	298	298	298	298	298	300	300	300	300	300
$^c\mathrm{N}_\mathrm{c}$	0	47	475	0	22	269	0	54	569	0	0	0	0	0
$^{ m w}{ m N}_q$	26265	26124	24840	31572	31401	29865	31572	29865	29865	10240	11008	10240	11008	11008
$^a\mathrm{N}_1$	350:88	350.88	350:88	150:150	150:150	150:150	150:150			256	128	128	256	128
$CaCl_{2}(M)$	0	0.1	1.0	0	0.1	1.0	0	0.1	1.0	0	0	0	0	0
NaCl (M)	0	0	0	0	0	0	0	0	0	0	0	0	0	0
force field for lipids $/$ ions	$Lipid17 / Dang^{??}$	Lipid17 / Dang???	Lipid17ecc / ECC-ions???	Lipid17ecc / ECC-ions???	Lipid17ecc / ECC-ions???	Berger? 17.	Berger [?] 18.	Berger? 19.	Berger? 20.	Berger [?] 21.				
lipid/counter-ions	POPC:POPG (4:1)	POPC:POPG (4:1)	POPC:POPG (4:1)	POPC:POPG (1:1)	POPC:POPG (1:1)	POPC:POPG (1:1)	POPC:POPG (1:1)	POPC:POPG (1:1)	POPC:POPG (1:1)	POPC	POPC:POPE (1:1)	POPC:DOPE (1:1)	DOPC	DOPC:DOPE (1:1)

 $[^]a\mathrm{Number}$ of lipid molecules with largest mole fraction

22. Citation and ion model for GROMOS-CKP?

23. Citation and description for "Berger" model?

24.Lipid17 POPC and POPC:POPG mixtures (https://doi.org/10.5281/zenodo.3241242 and https://doi.org/10.5281/zenodo.3237656) should be added after simulated with corrected dihedrals.

25. Upcoming Lipid17ecc with POPC:POPS (4:1) mixture simulations to be added.

 $[^]b\mathrm{Number}$ of water molecules

 $[^]c$ Number of additional cations

 $[^]d$ Simulation temperature e Total simulation time

fTime used for analysis

 $[^]g$ Reference for simulation files

POPE with additional NaCl 27. Simulation details by A. Peon.

POPG 28.Simulation details by Ollila.

POPG with additional NaCl 29. Simulation details by A. Peon.

POPC:POPE mixtures Data is available at.?? 300 K with v-rescale (tau=0.1 ps), 1 bar with PR semiisotropic (tau=4 ps, compressibility=4.5e-5 bar⁻¹), PME order 4 and space 0.12, recoulomb and rvdw 1.0, 128 lipids per leaflet, no ion 30.Full simulation details by Fuchs et al.

POPC:POPG mixture with additional calcium 31.Simulation details by J. Madsen.

POPC:POPG mixture with additional NaCl 32. Simulation details by A. Peon.

S2.2 CHARMM36ua

POPE Data is available at.? 33. Simulation details by T. Piggot.

S2.3 Slipids

POPE Data is available at. ? 34. Simulation details by T. Piggot.

POPE with additional NaCl 35. Simulation details by A. Peon. I have assumed that ion parameters are default Slipids, i.e., Aqvist, please correct if this is not true.

DPPE Data is available at.? 36.Simulation details by F. Favela.

POPG Data is available at.? 37.Simulation details by F. Favela. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not true.

POPG with additional NaCl 38. Simulation details by A. Peon. I have assumed that ion parameters are default Slipids, i.e., Åqvist, please correct if this is not true.

DPPG Data in 298 K is available at? and in 314 K at.? 39. Simulation details by F. Favela. I have assumed that ion parameters are default Slipids, i.e., Aqvist, please correct if this is not true.

POPC:POPG mixture with additional NaCl 40.Simulation details by A. Peon. I have assumed that ion parameters are default Slipids, i.e., Aqvist, please correct if this is not true.

S2.4 Berger

POPE Data is available at.?? 41.Simulation details by T. Piggot.

DOPE Data is available at.?? 42.Simulation details by T. Piggot.

POPC:POPE, POPC:DOPE and DOPC:DOPE mixtures Data is available at.?? 300 K with v-rescale (tau=0.1 ps), 1 bar with PR semiisotropic (tau=4 ps, compressibility=4.5e-5 bar⁻¹), PME order 4 and space 0.12, recoulomb and rvdw 1.0, 128 lipids per leaflet, no ion 43.Simulation details by Fuchs et al.

S2.5 GROMOS 43A1-S3

POPE Data is available at.? 44.Simulation details by T. Piggot.

S2.6 OPLS-UA

POPE Data is available at. [?] 45.Simulation details by T. Piggot.

POPE with vdW interaction in H Data is available at. [?] 46.Simulation details by T. Piggot.

S2.7 GROMOS-CKP and GROMOS-CKPM

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POPE Data is available at. <sup>?</sup> 47. Simulation details by T. Piggot.
DOPE Data is available at. <sup>?</sup> 48. Simulation details by T. Piggot.
DPPE Data is available at. <sup>?</sup> 49. Simulation details by T. Piggot.
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S2.8 Lipid17

S2.9 ECC-LIPID POPG

In ECC-lipid models, electronic continuum correction (ECC) is applied to implicitly include the missing electronic polarizability into the force field description. [?] In practise, this is implemented by scaling the charges and Lennard-Jones σ s of headgroup, glycerol backbone,

and carbonyl regions of Amber Lipid14/17 models are scaled by constant factors. Here, we follow the approach that previously improved ion binding to bilayers containing negatively charged PS lipids: PCC-POPC parameters (scaling factors f_q =0.8 and f_σ =0.89 applied to Lipid14 POPC parameters) were used for POPC and scaling factors of f_q =0.75 and f_σ =0.89 were applied to the charges and Lennard-Jones σ s of headgroup, glycerol backbone, and carbonyl regions of Amber Lipid17 POPG parameters. The Lipid17 parameters (described above) and initial configurations were taken from Ref. with the correct dihedral type, and the resulting parameters are available from Ref. CCC-ion parameters with the scaled charges, downloaded from bitbucket.org/hseara/ions/src/master/, were used in these simulations.

S3 R-PDLF and SDROSS experiments



Figure S3: (A) Chemical structure of POPE with the labeling of headgroup and glycerol backbone carbons. (B) INEPT spectra from POPE sample with the headgroup and glycerol backbone peaks labeled. (C) 2D R-PDLF spectra (D) Dipolar sliced from the 2D R-PDLF spectra with the resulting order parameters on top of figures. (E) Experimetal S-DROSS curves giving signs of the order parameters.

50.A, B etc. labels to be put in the figure.



Figure S4: (A) Chemical structure of POPG with the labeling of headgroup and glycerol backbone carbons. (B) INEPT spectra from POPG sample with the headgroup and glycerol backbone peaks labeled. (C) 2D R-PDLF spectra (D) Dipolar sliced from the 2D R-PDLF spectra with the resulting order parameters on top of figures. (E) Experimetal S-DROSS curves giving signs of the order parameters.

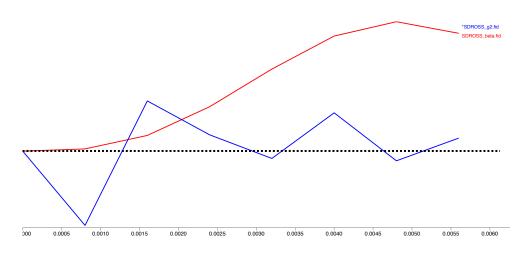


Figure S5: Simpson simulaton of S-DROSS curve of β -carbon of POPG.

S4 Changes of PG headroup order parameters upon addition of PC

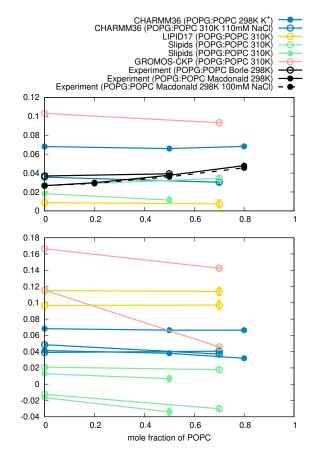


Figure S6: Modulation of PG lipid headgroup order parameters with the increasing amount of PC in lipid bilayer from experiments $^{?}$ and simulations with different force fields.

S5 Sodium binding to POPC simulations

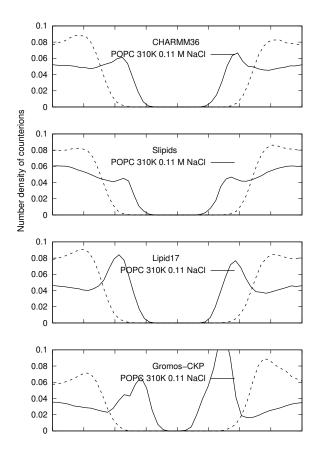


Figure S7: Sodium (solid line) and choride ion density profiles along membrane normal from different simulations with PC lipids.

51.Discussion about differences to the NMRlipids II to be discussed once we have the details on ions models.

S6 Calcium binding to POPC:POPG (4:1) mixtures

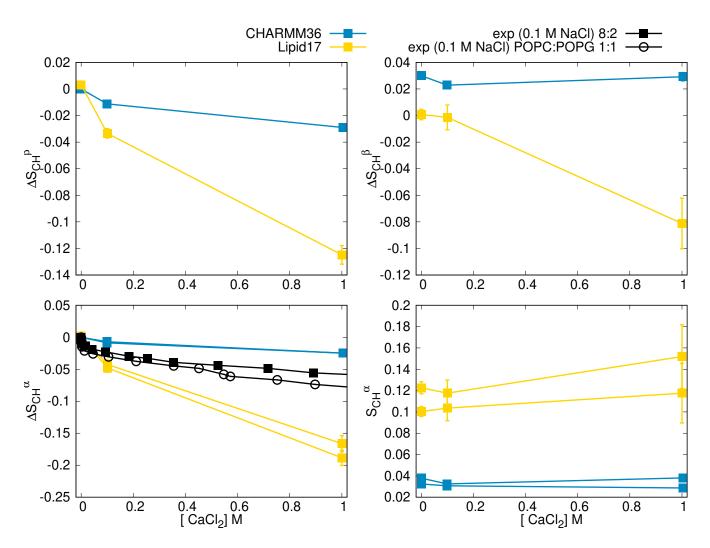


Figure S8: Modulation of headgroup order parameters of POPC (left) and POPG (right) in POPC:POPG (4:1) mixture upon addition of $CaCl_2$ in 298 K temperature from experiments? and simulations. The changes with respect to the systems without $CaCl_2$ are shown for other data than for the α -carbon of POPG for which experimental order parameter is not available.

52.Lipid17ecc data to be analyzed and added.

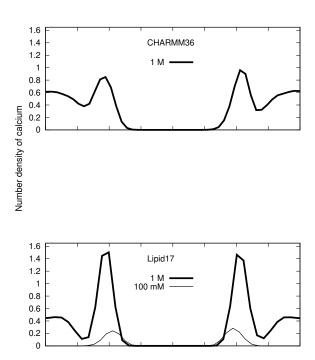


Figure S9: Calcium ion density profiles along membrane normal from simulations of POPC:POPG (4:1) mixtures with different force fields.