Supporting Information for

General Cluster Sorption Isotherm: Surface area determination

Christoph Buttersack

Institute of Chemical Technology, Leipzig University, Linnéstr. 3, 04103 Leipzig, Germany

Appeared in: **Physical Chemistry Chemical Physics** (2022)

Content

Table S1: Direct transformation of the 1-dimensional to the 2-dimensional GCSI. Data belonging to nitrogen sorption on alumina and differently modified silica (see Table 1). Data points taken from those inside the experimental pressure range. Q in g kg^{-1}

adsorbent	alumina		silica SH		silica ODM		silica	
temperature	77 K		77 K		77 K		77 K	
data points	1374		1406		1344		1411	
steepness	2860		1260		27		10100	
GCSI	2-dim		2-dim		2-dim		2-dim	
AICc	-12364		-6800		-8031		-12530	
R	0.99996		0.9994		0.9995		0.99998	
ln K_1	11.55	± 0.0075	10.209	± 0.032	6.178	± 0.020	12.498	± 0.00603
K	0.9894	± 0.0001	0.9541	± 0.0009	0.9606	± 0.0007	0.9615	± 0.00017
a	0.8142	± 0.0043	3.37	± 0.21	1.813	± 0.051	0.7683	± 0.00259
b	0.0569	± 0.0002	0.0769	± 0.0017	0.0579	± 0.0010	0.0737	± 0.00035
Q_1	0.2829	0.0009	1.059	0.017	0.920	0.013	0.9527	0.00330
$Q_{m\,GCSI}$	1.63		3.62		4.87		4.57	
Table 1:	1.64		3.64		4.17		4.60	

Table S2: Direct transformation of the 1-dimensional to the 2-dimensional GCSI. Data belonging to the sorption of argon, water, neo-pentane, and n-pentane on macroporous silica (see Table 2). Data points taken from those inside the experimental pressure range. Q in g kg^{-1}

adsorbate	argon		water		neo-pentane		n-pentane	
temperature	87 K		303 K		273 K		298 K	
data points	1305		1090		918		1032	
steepness	89.5		17.1		5.70		6.25	
GCSI	2-dim		2-dim		2-dim		2-dim	
AICc	-7373		-7767		-33174		-8515	
R	0.9995		0.99995		1.0000000		0.99991	
ln K_1	7.283	± 0.020	3.4019	± 0.0083	3.0725	± 3E-08	2.8664	± 0.0302
K	0.95501	± 0.00072	0.9807	± 0.0003	0.9609	± 4E-09	0.9827	± 0.0031
a	3.12	± 0.16	>6	±	-0.0048	± 8E-08	>6	±
b	0.0797	± 0.00137	0.4628	± 0.0056	0.2406	± 2E-08	0.1353	± 0.0043
Q_1	1.085		7.84	0.0378	10.95	4E-07	7.47	0.1939
$Q_{m\,GCSI}$	4.15		12.26		30.83		24.72	
Table 2:	3.93		12.26		30.83		24.77	

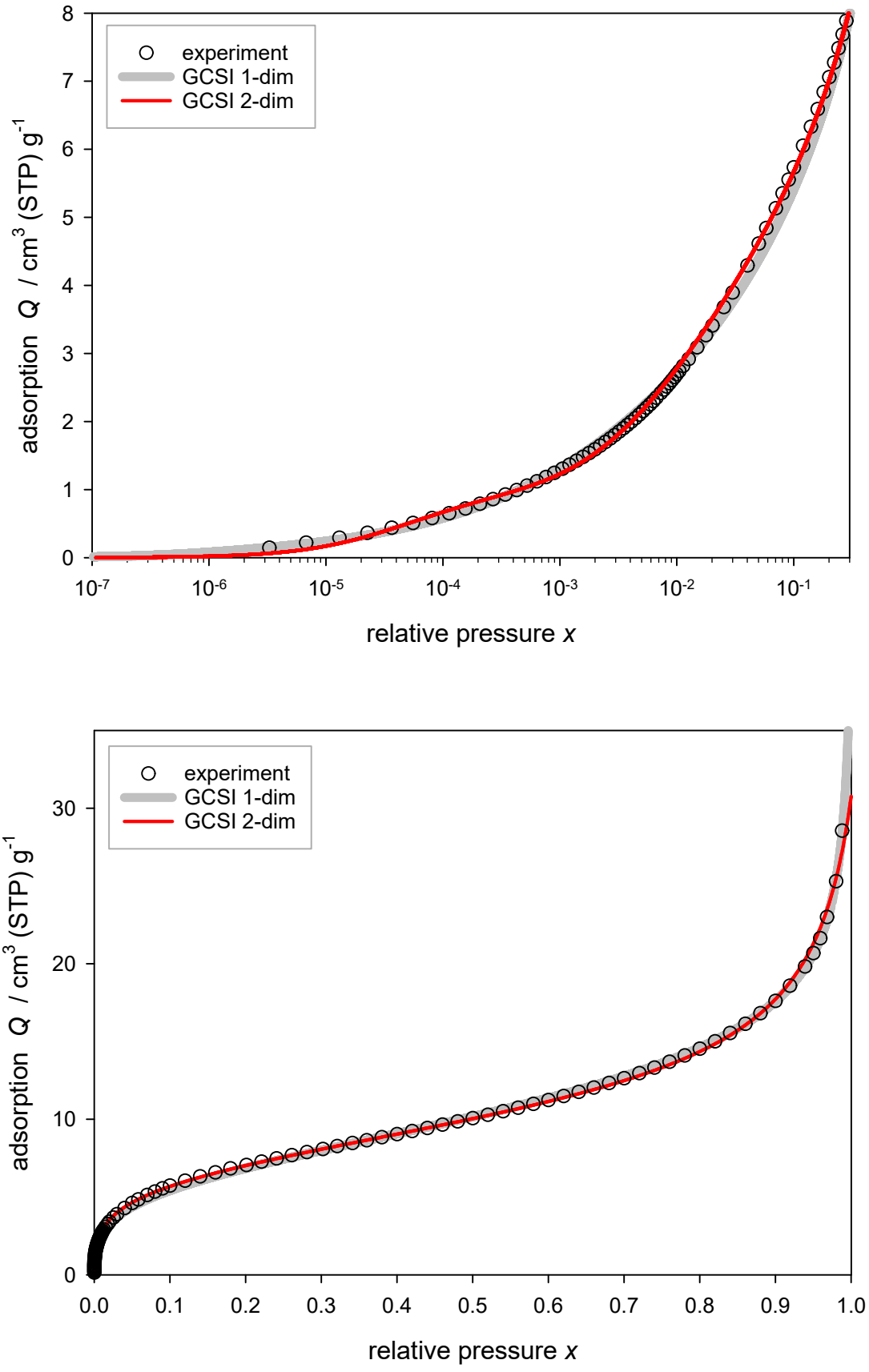

Figure S1: 1-and 2-dimensional GCSI applied to nitrogen sorption on SH functionalized macroporous silica at 77 K. Data from literature (see Table 1).

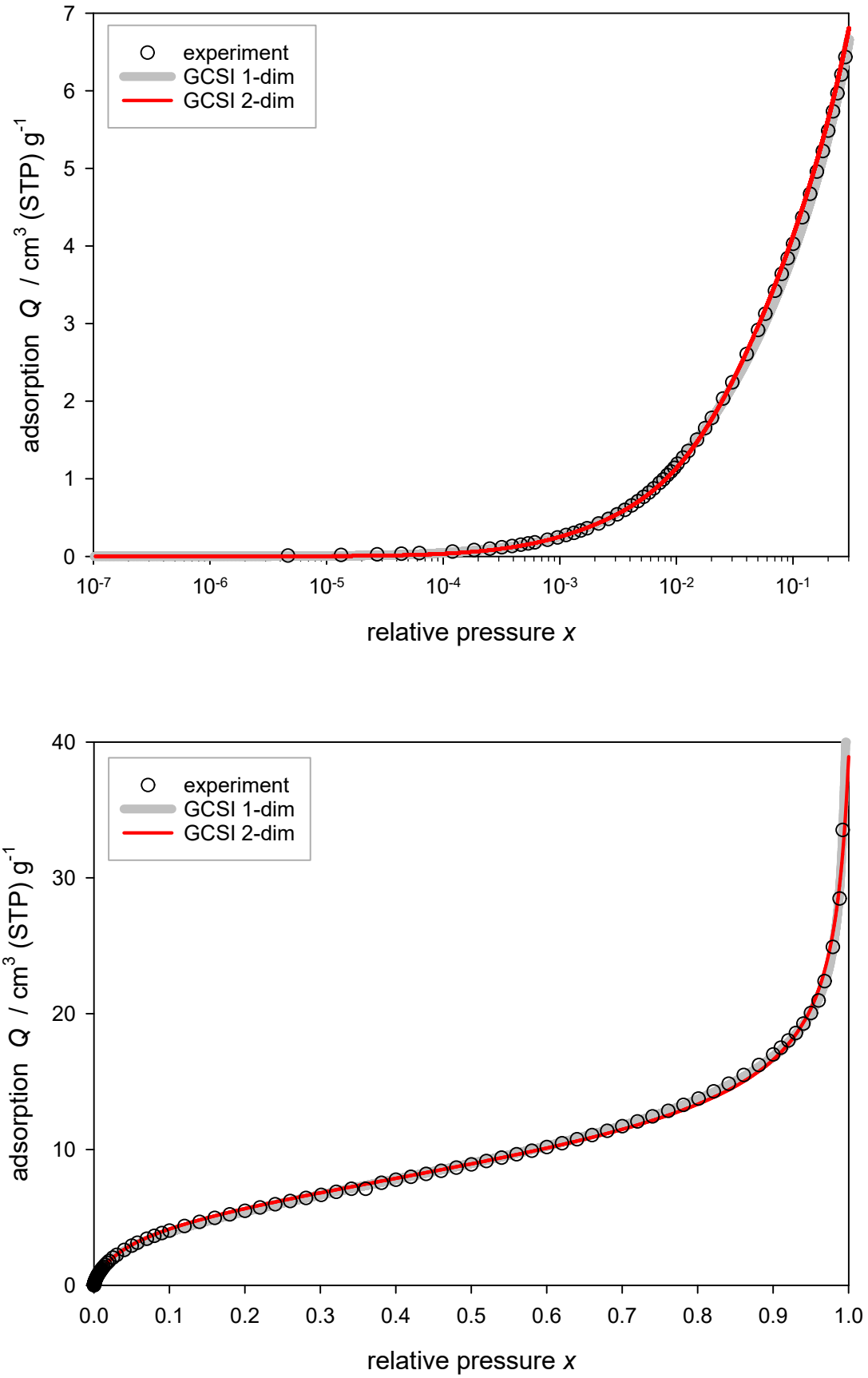

Figure S2: 1-and 2-dimensional GCSI applied to nitrogen sorption at 77 K on macroporous silica functionalized with octyldimetyl residues (ODM). Data from literature (see Table 1).

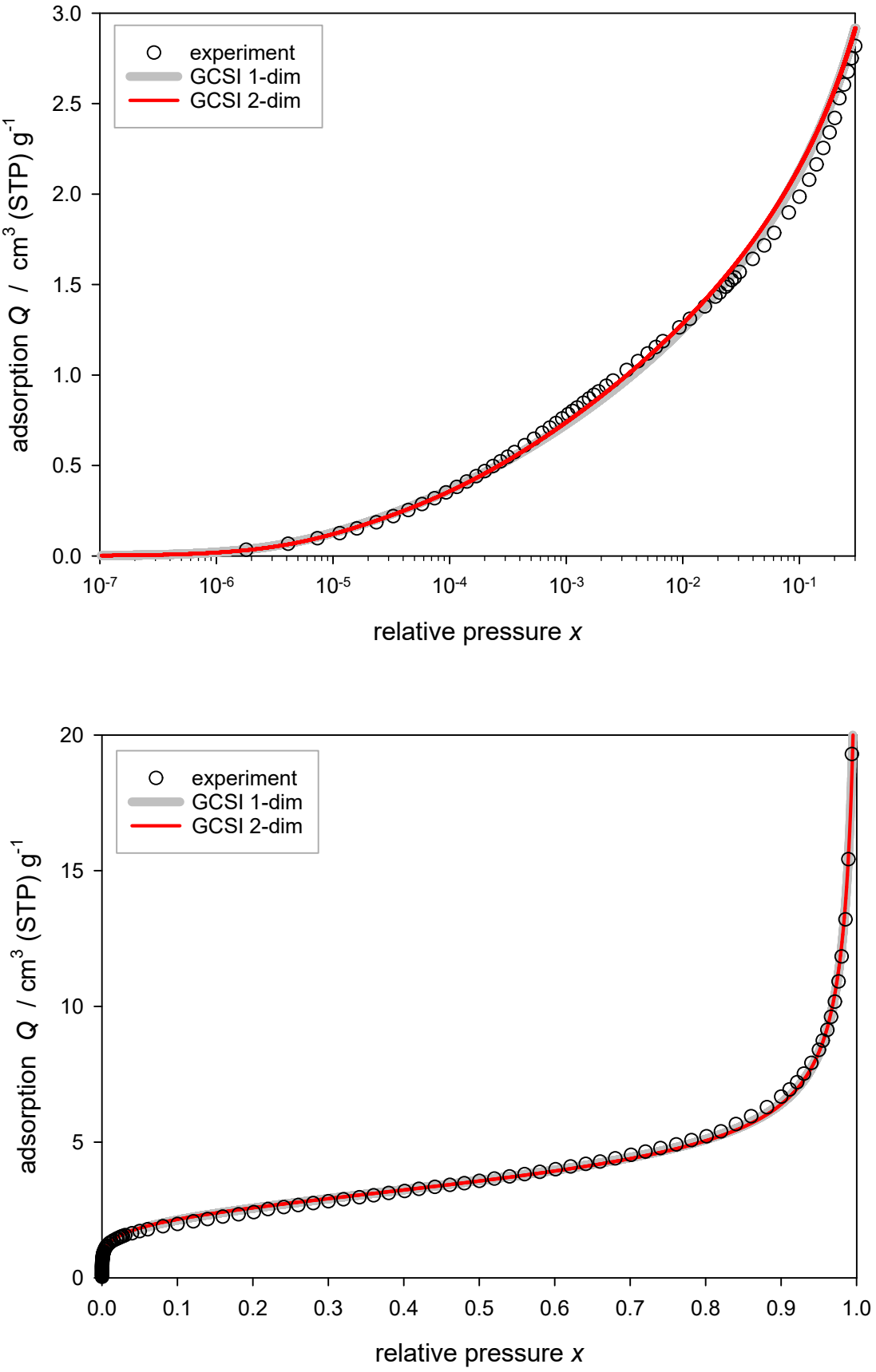

Figure S3: 1-and 2-dimensional GCSI applied to nitrogen sorption at 77 K on macroporous alumina. Data from literature (see Table 1).

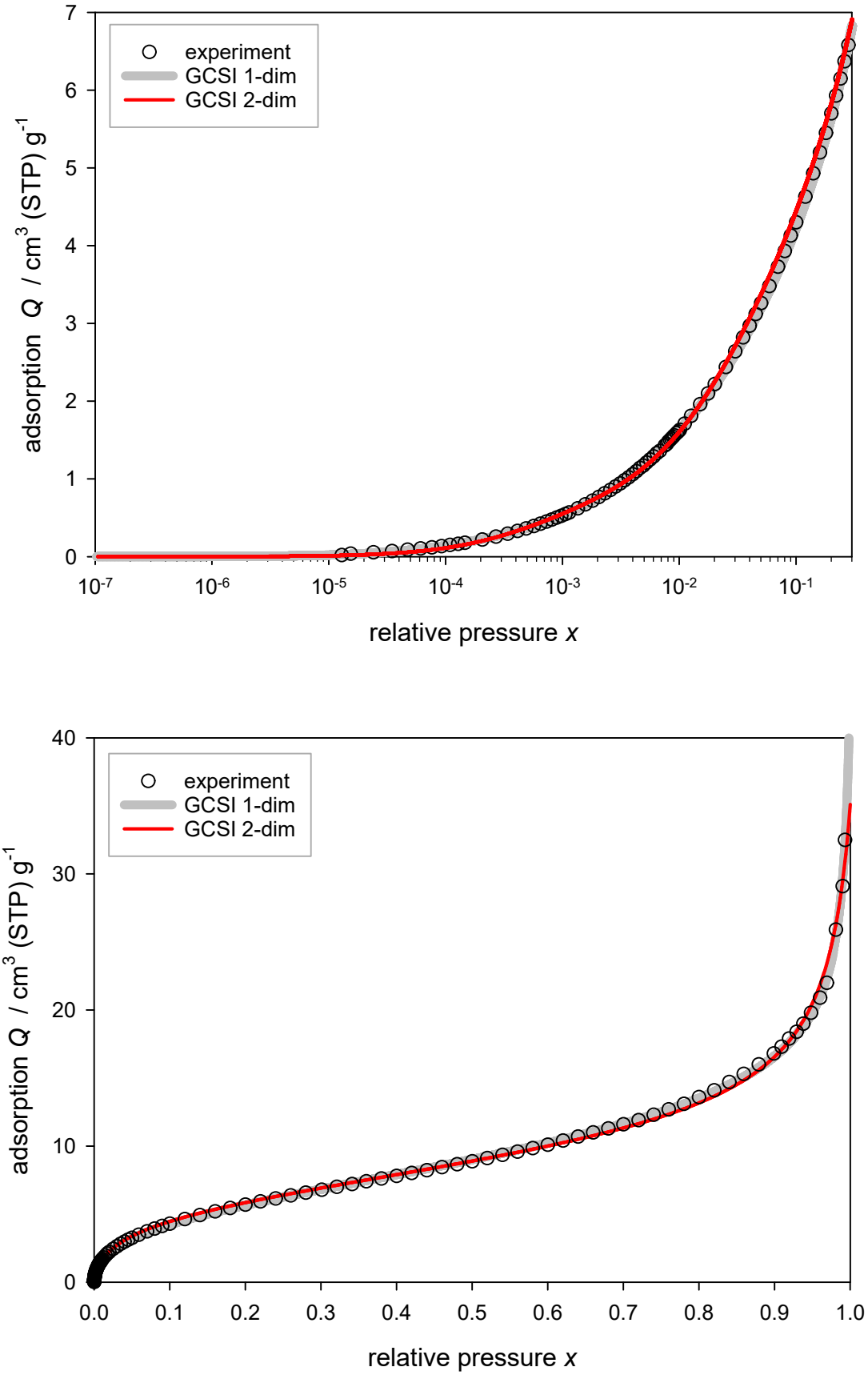

Figure S4: 1-and 2-dimensional GCSI applied on argon sorption on macroporous silica at 87 K. Data from literature (see Table 2).

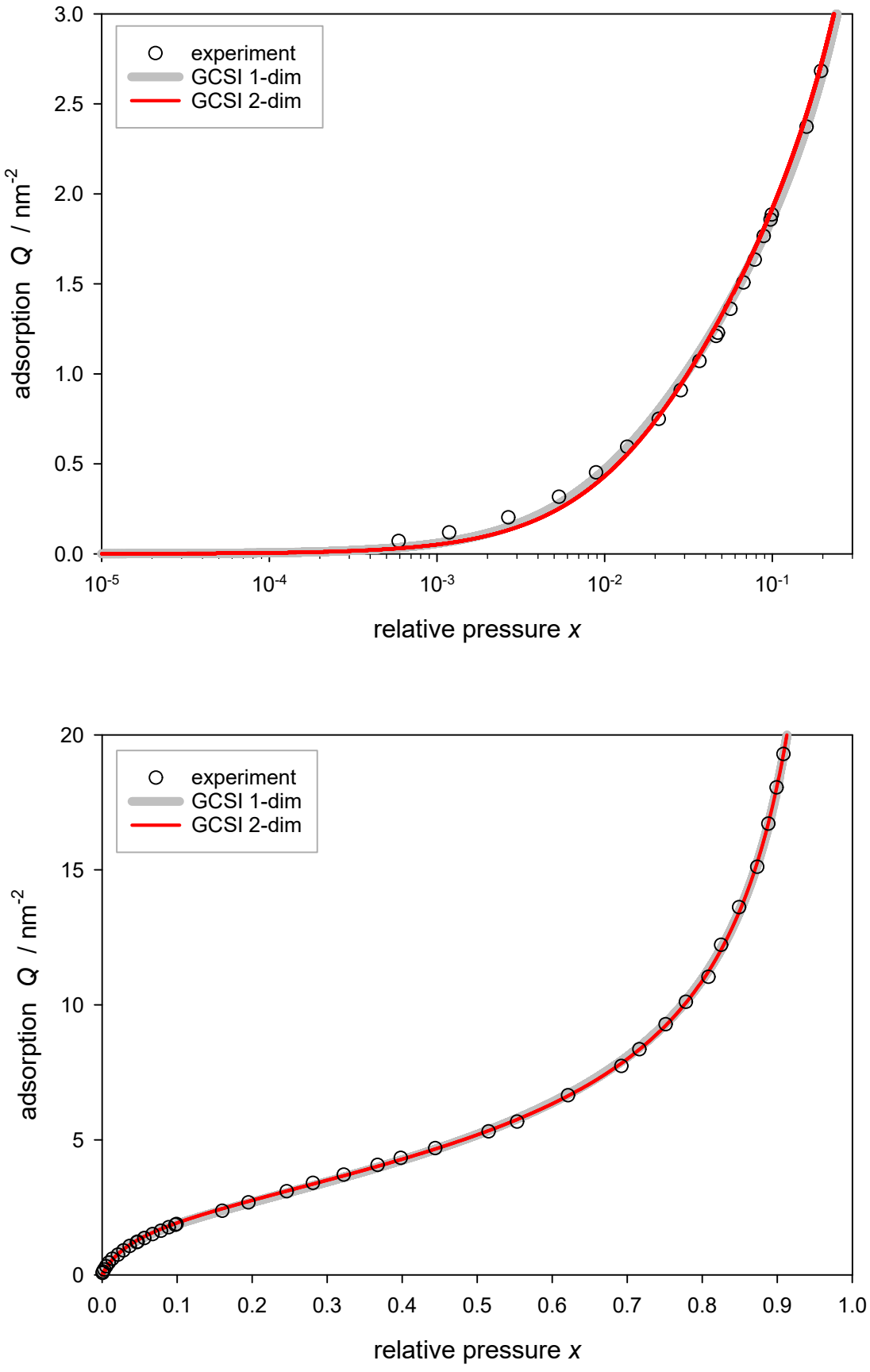

Figure S5: 1-and 2-dimensional GCSI applied on water sorption on macroporous silica at 303 K. Data from literature (see Table 2). Dimension of *Q*: see original literature and ref. 13.

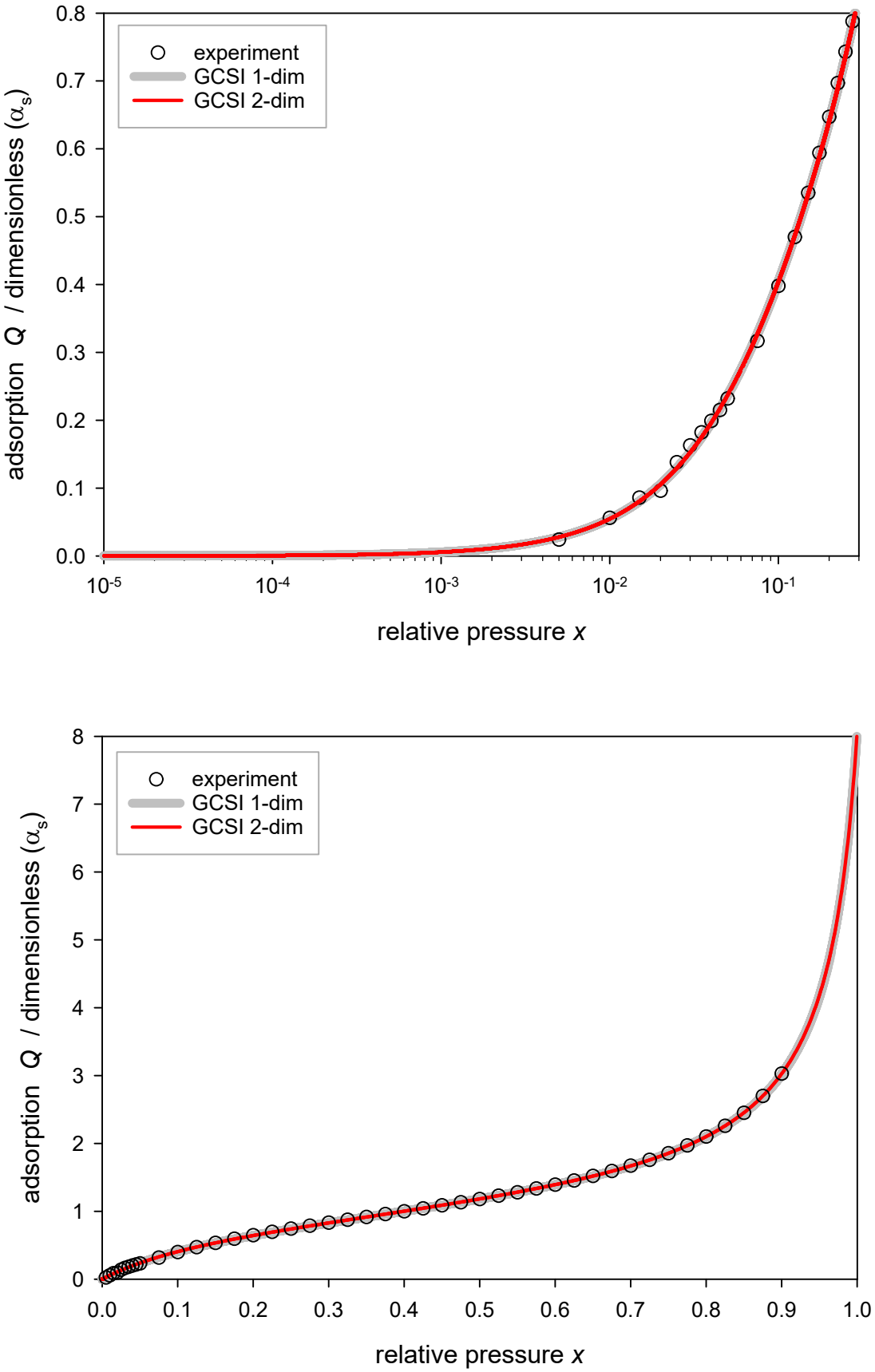

Figure S6: 1-and 2-dimensional GCSI applied on neo-pentane sorption on macroporous silica at 273 K. Data from literature (see Table 2). Dimension of *Q*: see original literature and ref 13.

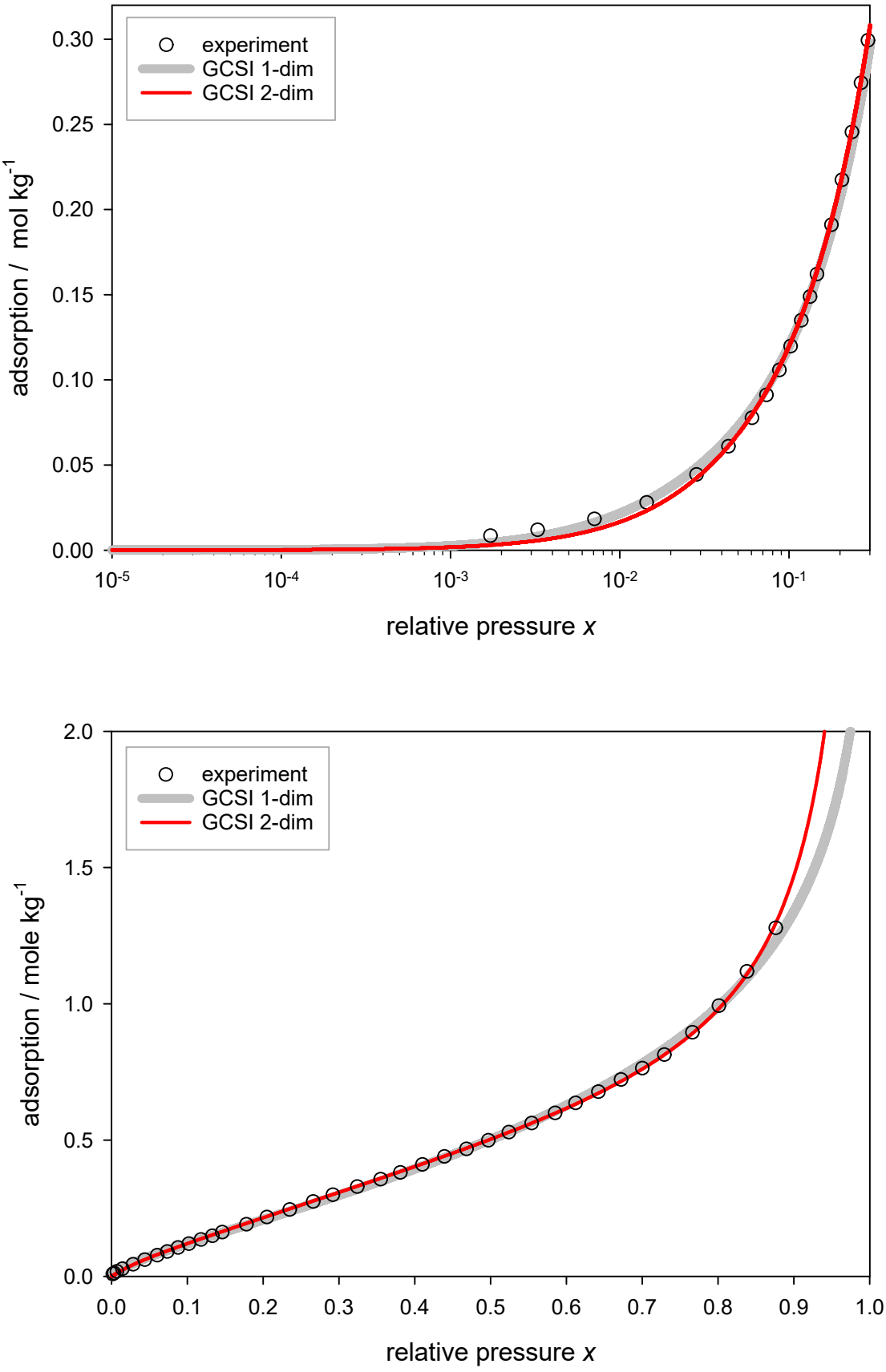

Figure S7: 1-and 2-dimensional GCSI applied on n-pentane sorption on macroporous silica at 298 K. Data from literature (see Table 2). For the 1-dimensional case *K* is arbitrarily set to 0.968.

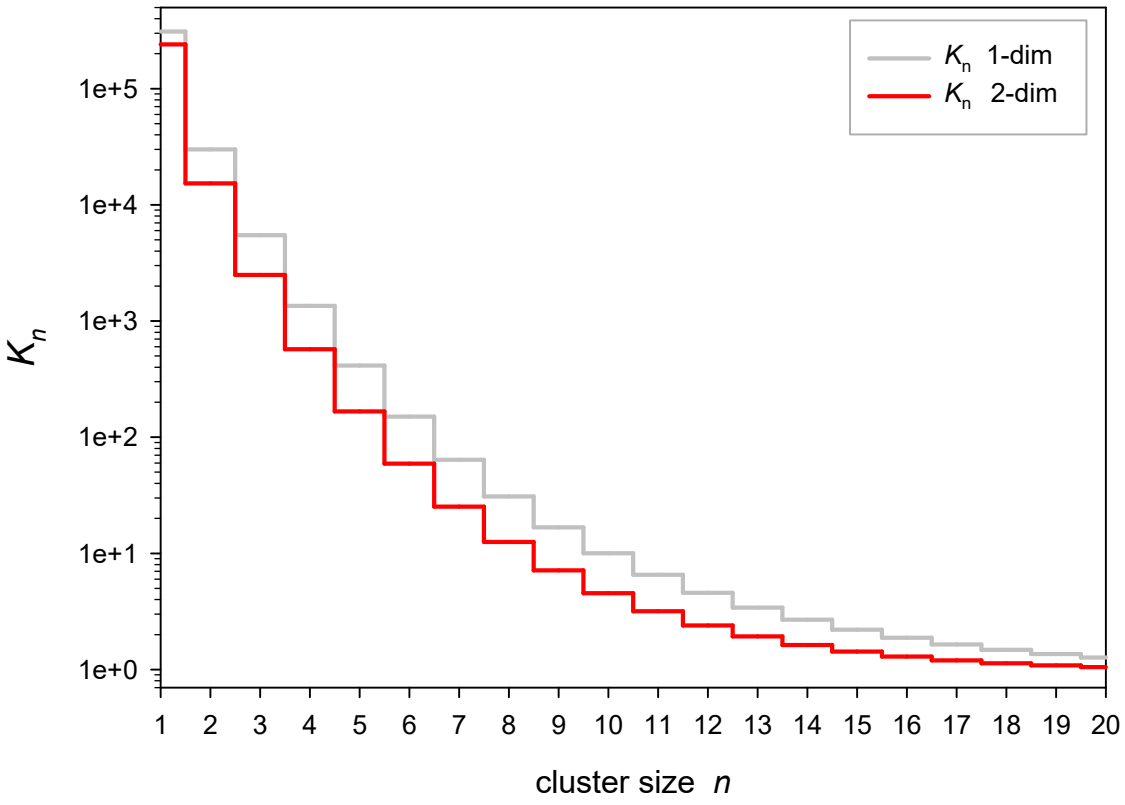

Figure S8: Plot of K_n versus cluster size for adsorption of nitrogen on macroporous silica at 77 K

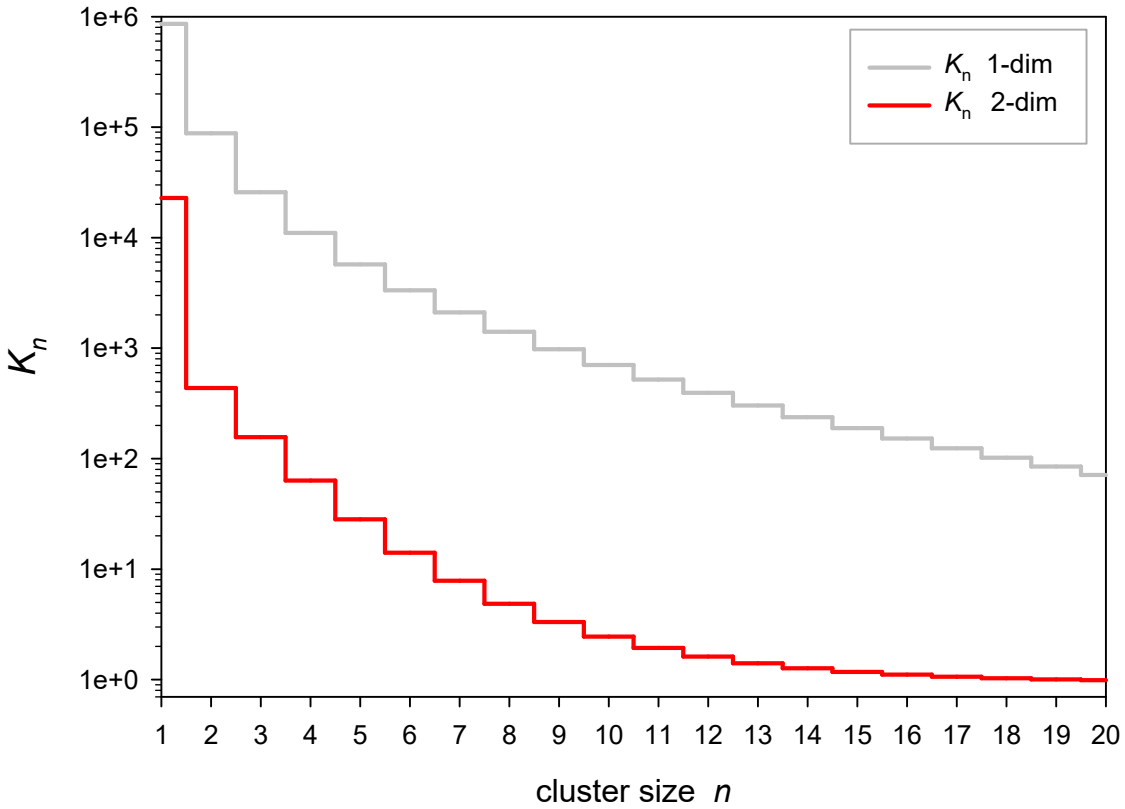

Figure S9: Plot of K_n versus cluster size for adsorption of nitrogen at 77 K on macroporous silica functionalized with SH groups.

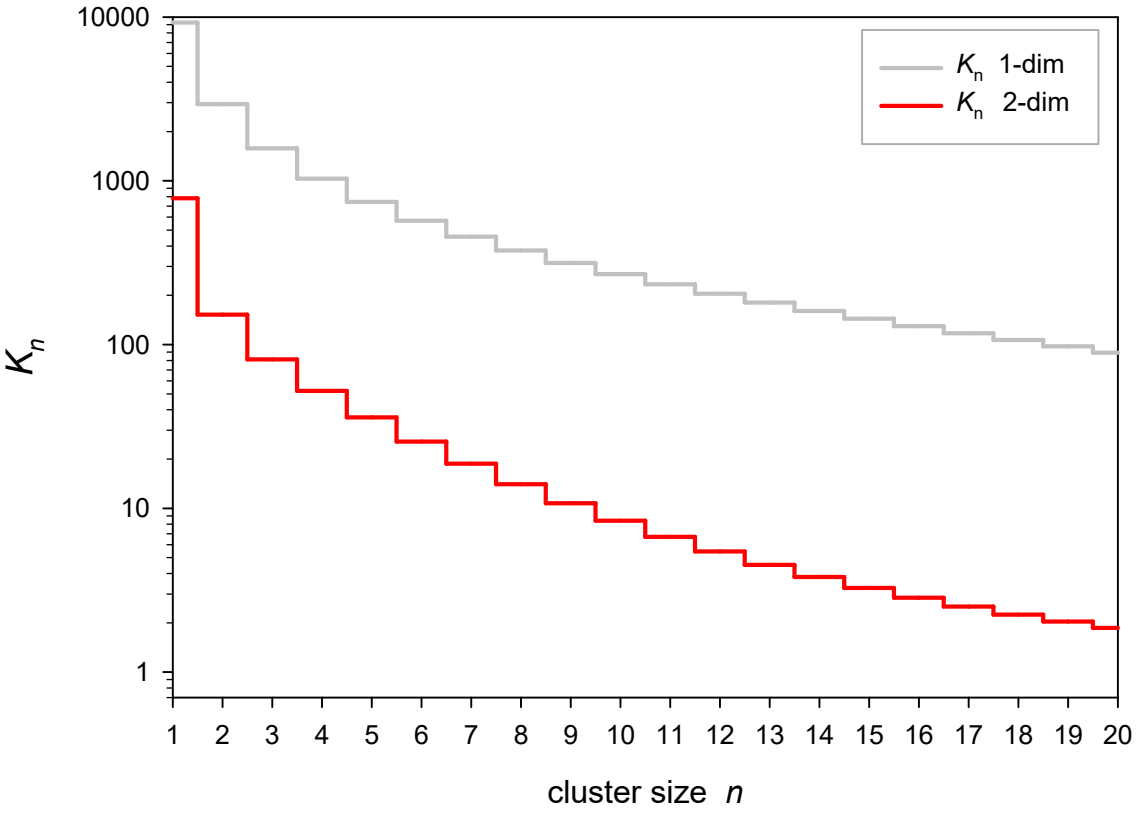

Figure S10: Plot of K_n versus cluster size for adsorption of nitrogen at 77 K on macroporous silica functionalized with octyldimethyl groups (ODM).

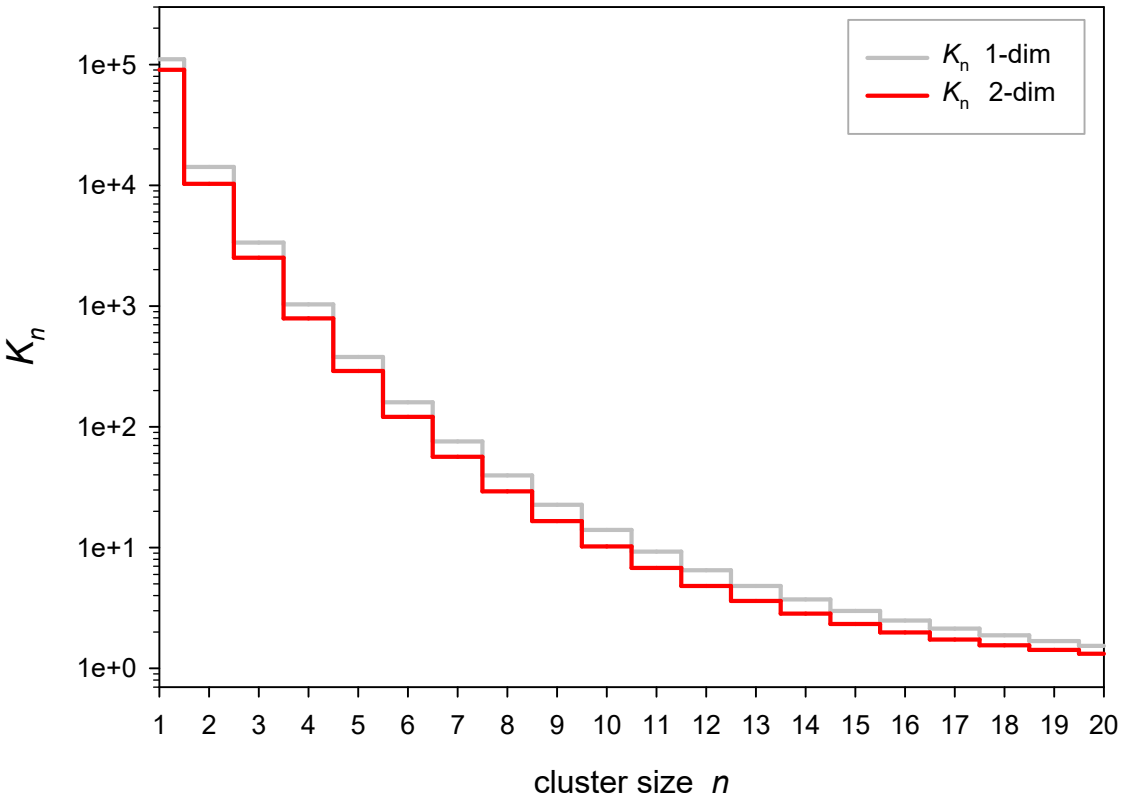

Figure S11: Plot of K_n versus cluster size for adsorption of nitrogen at 77 K on macroporous alumina.

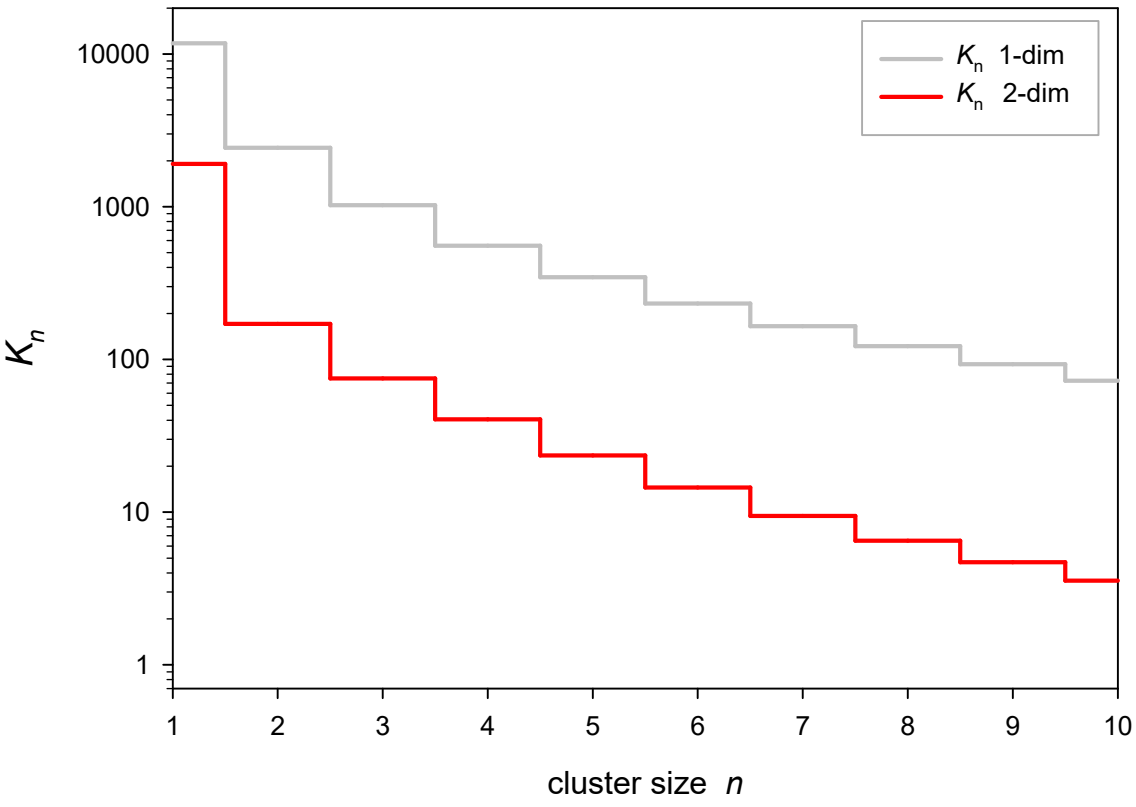

Figure S12: Plot of K_n versus cluster size for adsorption of argon at 87 K on macroporous alumina

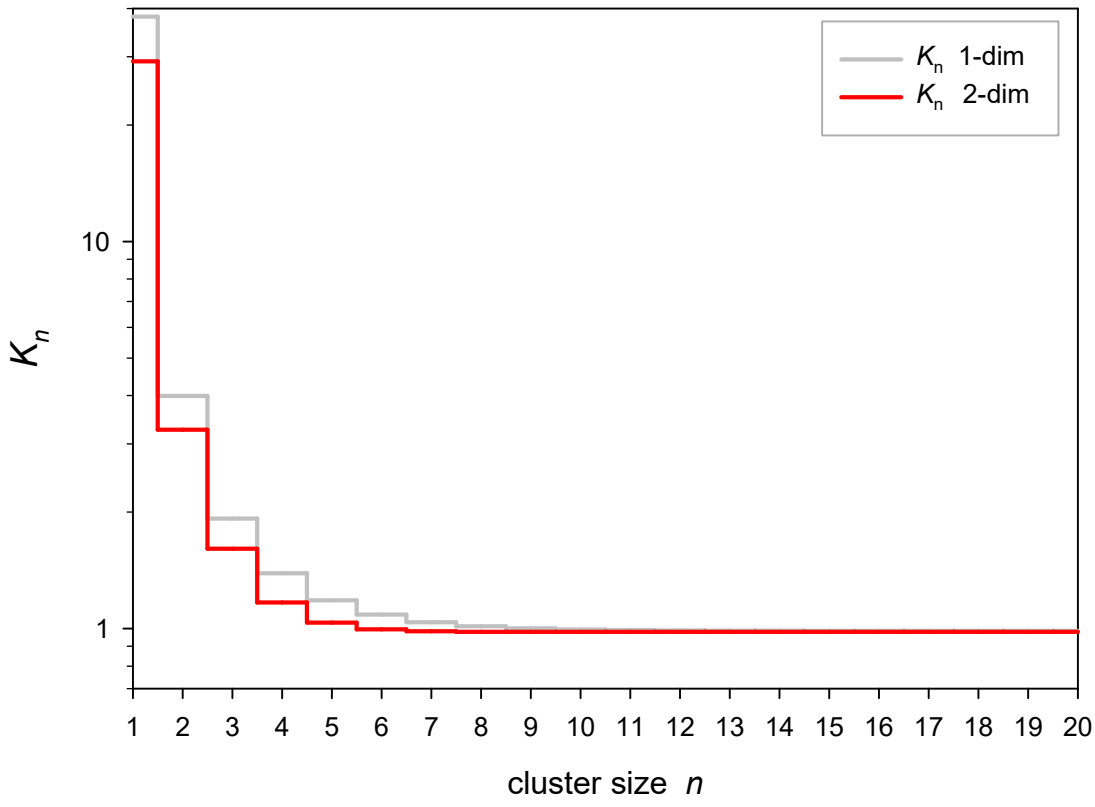

Figure S13: Plot of K_n versus cluster size for adsorption of water at 303 K on macroporous silica.

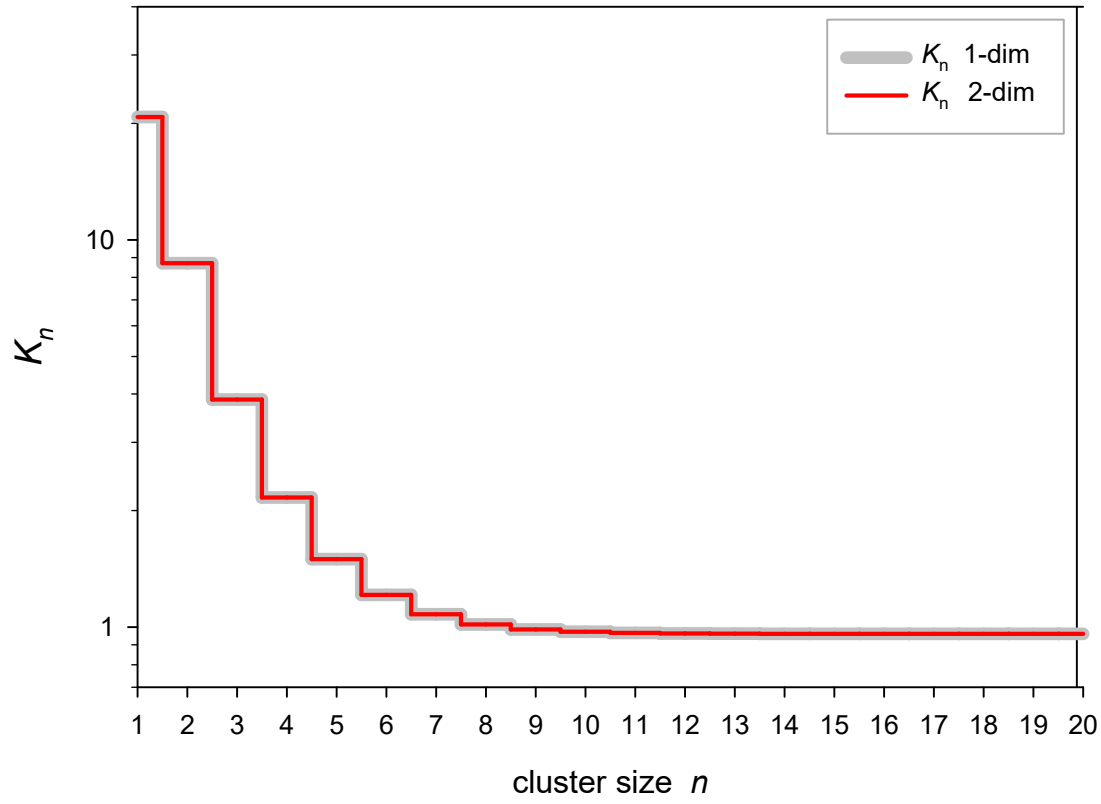

Figure S14: Plot of K_n versus cluster size for adsorption of neo-pentane at 273 K on macroporous silica.

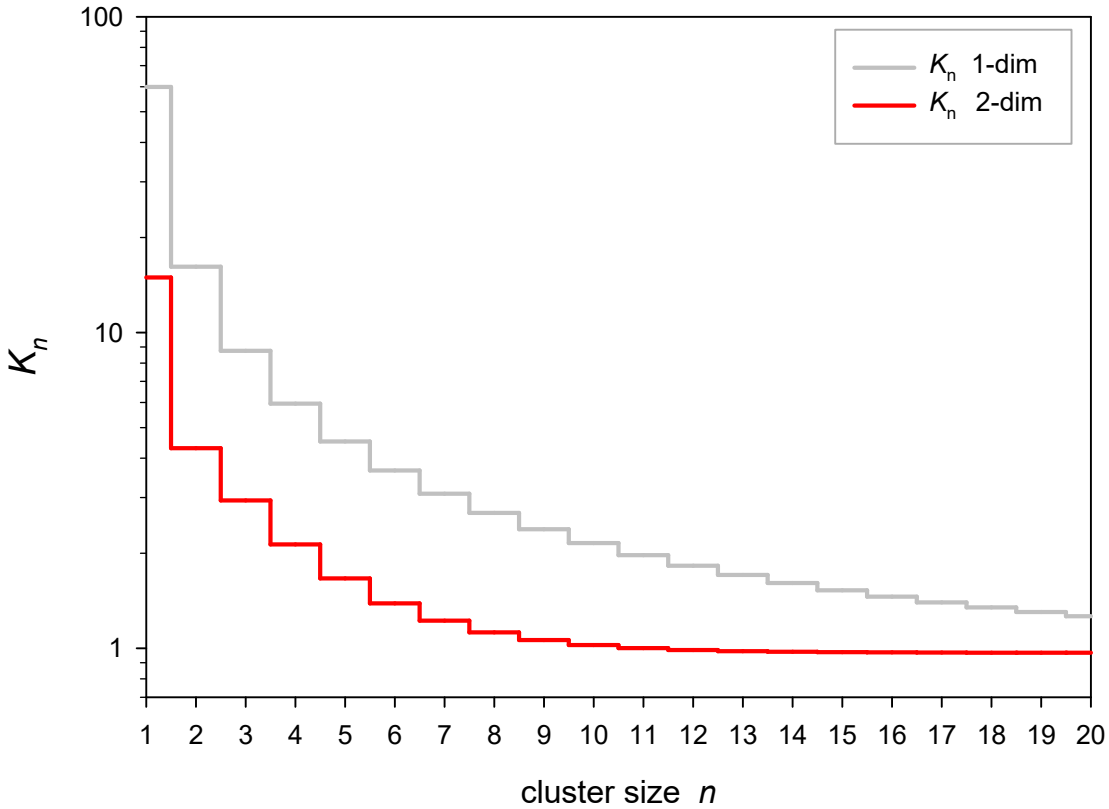

Figure S15: Plot of K_n versus cluster size for adsorption of n-pentane at 298 K on macroporous silica. For the 1-dimensional case K is arbitrarily set to 0.968.

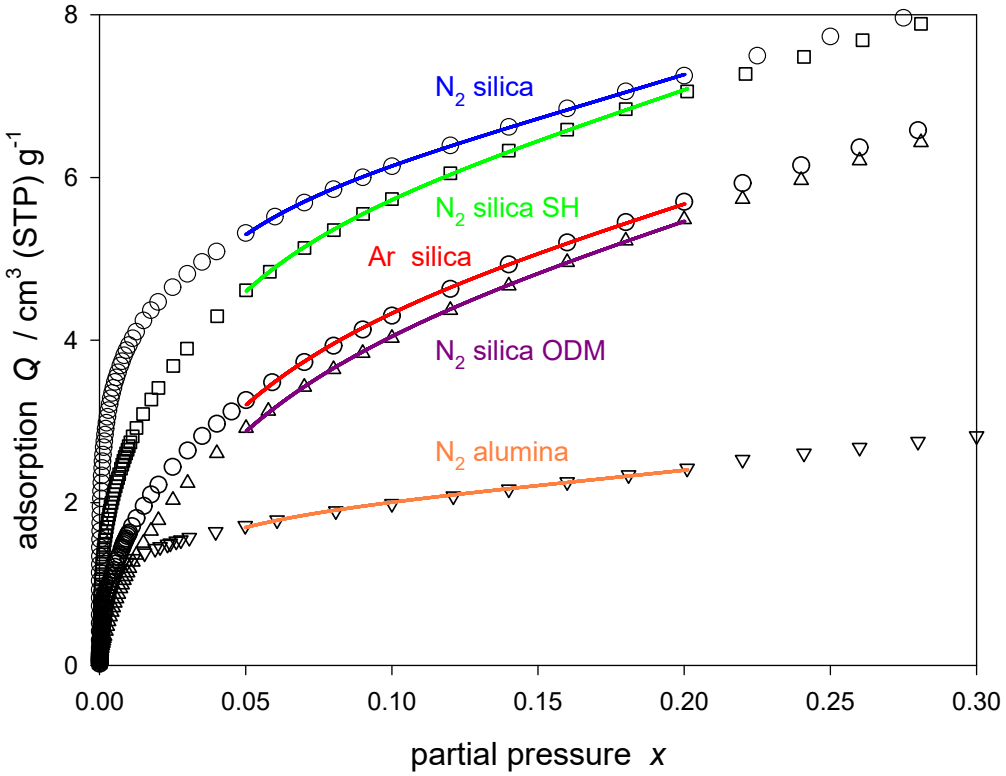

Figure S16: Monolayer detection by BET ($0.05 < x < 0.2$). Q_{BET} in $cm^3\ g^{-1}$:
N_2 silica: 6.064±0.009, Ar silica: 6.147±0.012, N_2 silica SH: 6.147± 0.012, N_2 silica ODM: 5.200±0.023, N_2 alumina: 2.022±0.011

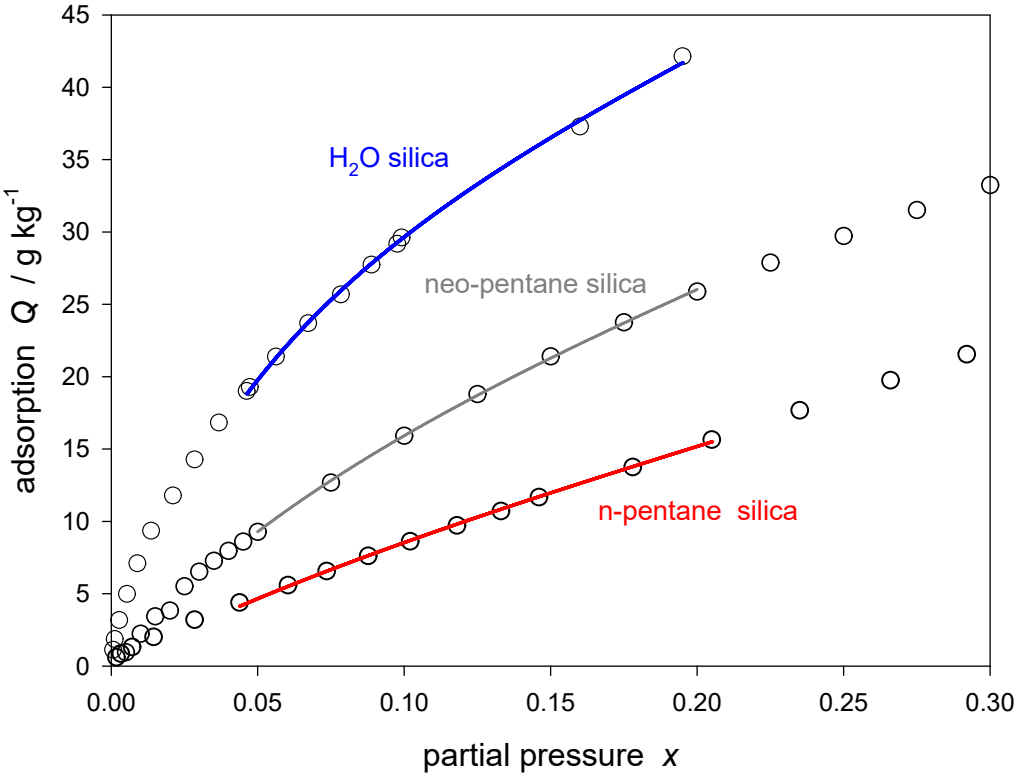

Figure S17: Monolayer detection by BET ($0.05 < x < 0.2$). Q_{BET} in $g\ kg^{-1}$:
water silica: 11.8±0.1, neo-pentane silica: 32.7±0.4, n-pentane silica: 22.7±0.5

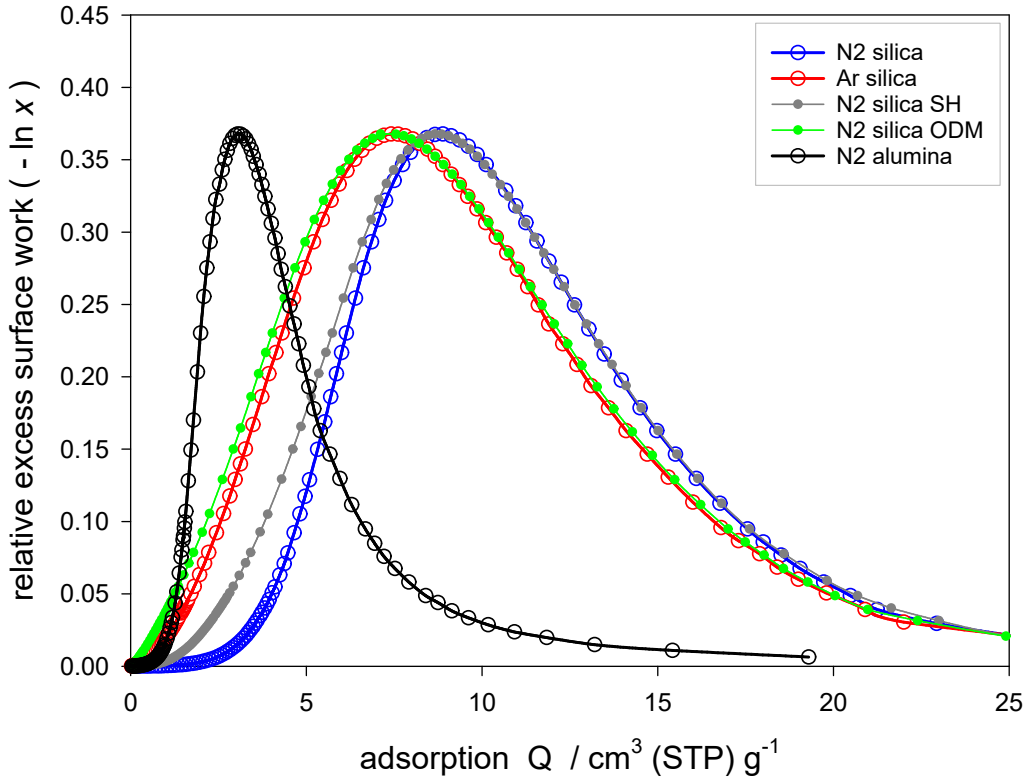

Figure S18: Monolayer detection by excess surface work (ESW). Q_{ESW} in $cm^3\ g^{-1}$: N_2 silica: 8.944±0.007, Ar silica: 7.5872±0.0008, N_2 silica SH: 8.832± 0.005, N_2 silica ODM: 7.532±0.006, N_2 alumina: 3.114±0.004

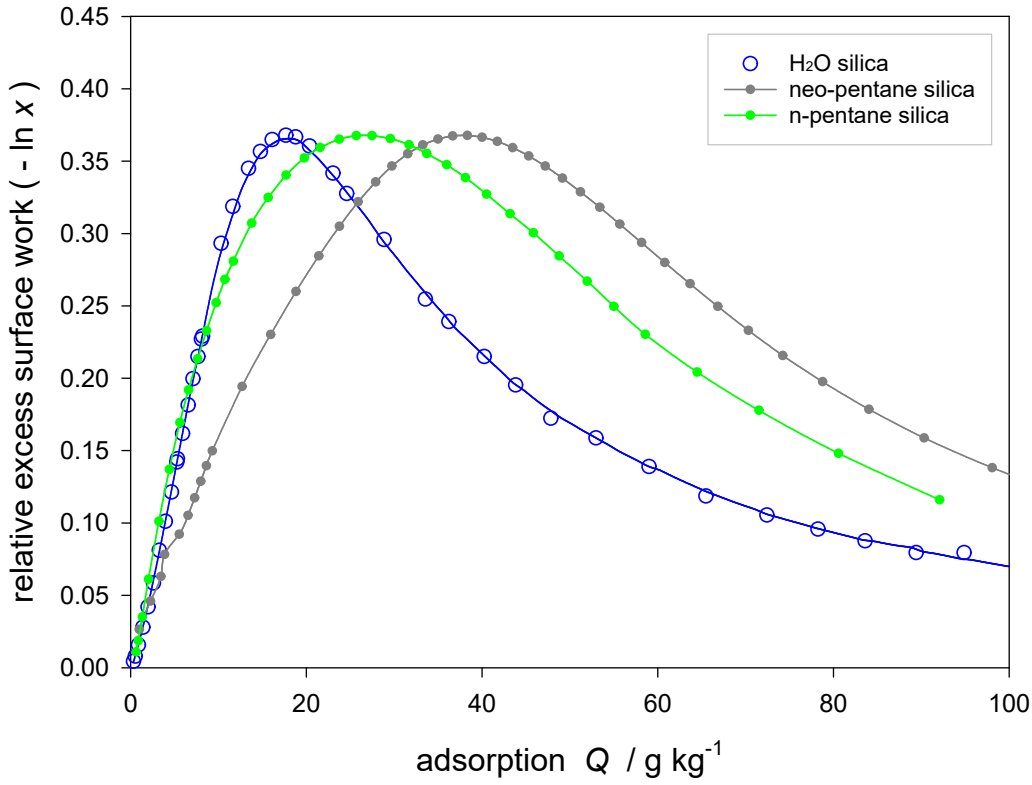

Figure S19: Monolayer detection by excess surface work (ESW). Q_{BET} in $g\ kg^{-1}$: water silica: 18.5±0.1, neo-pentane silica: 38.77±0.06, n-pentane silica: 27.7±0.2

Program in R for nonlinear regression with the 2-dimensional general cluster sorption isotherm, version for type II isotherms

In addition to the programs in R for the 1-dimensional GCSI to be find in the supporting information of ref. [13], the program for the 2-dimensional GCSI is appended in the present SI. It yields the results E_1 = ln K_1, K, a, b, Q_1, and . For data input an excel file due to the following example has to be used:

rel pressure	adsorbed	estimated	parameters
5.55E-07	0.109	K1	150000
1.23E-06	0.210	K	0.95
2.38E-06	0.310	a	-0.18
4.18E-06	0.420	b	-0.055
6.74E-06	0.520	Q1	0.65
1.02E-05	0.620		
1.49E-05	0.730		
2.10E-05	0.830		
ect.	ect.		

Note: compared to the equations in the article the sign of a and b is inverted! Estimated start parameters are the starting values for non-linear regression. It is indicated within the running program if the limits if the parameters are reached. In that case the limits and perhaps some steps in the program have to be changed. The series of expansion (theoretically up to infinite) is terminated at β = 1000.

The program appends the results into the input file as new excel sheets concerning the calculated isotherm, calculated values of K_n and C_n ($1 \le n \le 1000$), and the optimized parameters including the correlation coefficient R, the value of the corrected Akaike information criterion $AICc$ [13] and the calculated monolayer adsorption in the dimension used for Q_1 in the input file.