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Articles

Henry's Law Constants of Selected Aromatic Hydrocarbons, Alcohols, and Ketones

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The Henry's law constants (air−water partition coefficients) are reported for 18 hydrocarbons (polynuclear aromatic and chlorinated benzenes), two alcohols, and three ketones at 25 °C determined by an equilibrium gas stripping method.

Introduction

The Henry's law constant or air−water partition coefficient is an important physical−chemical property of an organic chemical with applications in environmental and health sciences and in chemical analysis and processing. For example, it influences the rate of evaporation of organic solutes from water and soils, it determines the feasibility of head space and purge and trap analysis and it controls the effectiveness of gas stripping operations which are used to remove volatile organic solutes from water (Mackay, 1991). Mackay and Shiu (1981) have reviewed methods of determining, estimating, and expressing these coefficients and have compiled data for a variety of organic chemicals of environmental interest.

Briefly, the Henry's law constant H can be expressed as the ratio of partial vapor pressure and concentration of a solute in a dilute aqueous solution, i.e., (P/x) where P is the solute's partial pressure in the vapor phase and x is its mole fraction in the aqueous solution; or as P/C where C is the solute concentration in solution. Alternatively, it may be expressed as the air−water partition coefficient K_{AW} or C_A/C_W with identical concentration units in air, C_A, and water, C_W, e.g., $g{\cdot}m^{-3}$ or $mol{\cdot}m^{-3}$. Applying the ideal gas law shows that K_{AW} is H/RT where R is the gas constant and T is absolute temperature. H is often estimated as the ratio of solute vapor pressure P^S to aqueous solubility C^S, but this procedure fails for solutes which are in a supercritical state (e.g., methane) or have substantial solubilities in, or even miscibility with, water (e.g., alcohols). Experimental determination is then necessary.

Several techniques have been proposed in addition to the obvious direct determination of C_A and C_W. Mackay et al. (1979) developed an equilibrium gas stripping method, which is used here. The equilibrium partitioning in closed system (EPICS) method of Lincoff and Gossett (1984) and Gossett (1987) is particularly suitable for volatile solutes. A wetted-wall column technique has been described by Fendinger and Glotfelty (1988, 1990). Hussam and Carr (1985) have described a head space method, and Orbey and Sandler (1991) and Tse et al. (1992) have reported a gas chromatographic method. Compilations of data and correlations have been presented by Mackay et al. (1979), Mackay and Shiu (1981), Shiu and Mackay (1986), Suntio et al. (1988), Ashworth et al. (1988), Nirmalakhandan and Speece (1988), Yaws et al. (1991), and Meylan and Howard (1991).

In this study we report the experimental determination of Henry's law constants of 23 chemicals with a wide range in structure and properties at 25 °C by the equilibrium gas stripping method (Mackay et al., 1979). Air is bubbled at a known flow rate through a mixed water column containing the solute of interest, the exit gas achieving equilibrium with the water. By measuring the decrease in water concentration the Henry's law constant can be deduced from a mass balance.

Specifically, the decay in concentration of a solute from an aqueous solution in a vessel can be shown to be first order given by

$$\ln(C_W/C_{WO}) = -GK_{AW}t/V_W$$

where C_W and C_{WO} are the concentrations at time t and initially, G is the gas flow rate and V_W is the volume of water. A semilogarithmic plot of concentration in water as a function of time gives a slope of GK_{AW}/V_W from which K_{AW} can be estimated. This approach requires only that the concentration determinations be precise, not necessarily accurate.

Experimental Section

Chemicals. All hydrocarbons (of the highest commercially available quality) were obtained from Aldrich Chemical Co. and were used without further purification. Spectrograde cyclohexane was obtained from Caledon Laboratories, Georgetown, Ontario. Double distilled water was used in all experiments.

Apparatus. The apparatus for measuring the Henry's law constant has been described in detail by Mackay et al. (1979). Nitrogen was introduced through a sintered glass disk into the bottom of a glass stripping vessel thermostated at 25 °C filled with the aqueous solution of interest. Originally, the glass vessel was designed to contain 4.5 L of liquid so that the total liquid volume change would be insignificant with larger samples drawn for liquid extraction in order to increase the detection limit. The use of a smaller volume of 1 L was later found to be advantageous since it shortened the duration of the experiment. In both cases, the system was maintained at (25 ± 1) °C by Neslab

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Table 1. Henry's Law Constants for Selected Organic Compounds at 20−25 °C

compound	vap pres P^S/Pa[a]	solubility C^S/mol·m^{-3} [a]	H_{calc}/Pa·m^3·mol^{-1} P^S/C^S	H_{exptl}/Pa·m^3·mol^{-1} (this work)	H(lit.)/Pa·m^3·mol^{-1}	method	ref
naphthalene	10.4	0.242	43.01	44.6 ± 1.88	48.94	gas stripping	Mackay et al., 1979
					56	gas stripping	Southworth, 1979
					36.6	20 °C, EPICS[b]	Yurteri et al., 1987
					74.4	wetted-wall column	Fendinger and Glotfelty, 1990
					42.5	gas stripping	Alaee et al., 1996
1-methylnaphthalene	8.84	0.197	44.90	24.3 ± 1.80	26.3	gas stripping	Mackay et al., 1979
					62.0	wetted-wall column	Fendinger and Glotfelty, 1990
2-methylnaphthalene	9.0	0.176	51.20		32.2	wetted-wall column	Fendinger and Glotfelty, 1990
					20265	26 °C, EPICS, see text	Hansen et al., 1993
1,5-dimethylnaphthalene		0.0198		36.3 ± 1.54			
biphenyl	1.3	0.0454	28.64	31.2 ± 1.26	41.34	gas stripping	Mackay et al., 1979
					19.6	wetted-wall column	Fendinger and Glotfelty, 1990
acenaphthylene	0.9	0.107	8.40		11.55	gas stripping	Warner et al., 1987
acenaphthene	0.3	0.0246	12.17	16.2 ± 0.61	14.79	gas stripping	Mackay et al., 1979
					24.42	gas stripping	Warner et al., 1987
fluorene	0.09	0.0114	7.87	9.75 ± 0.23	11.86	gas stripping	Warner et al., 1987
					6.45	wetted-wall column	Fendinger and Glotfelty, 1990
phenanthrene	0.02	0.00617	3.24	3.61 ± 0.10	3.98	gas stripping	Mackay et al., 1979
					5.55	gas stripping	Southworth, 1979
					2.38	wetted-wall column	Fendinger and Glotfelty, 1990
					4.68	gas stripping	Alaee et al., 1996
anthracene	0.001	0.00025	3.96	7.66 ± 0.91	6.59	gas stripping	Southworth, 1979
					7.40	gas stripping	Mackay and Shiu, 1981
					7.40	gas stripping	Mackay et al., 1982
					1.96	wetted-wall column	Fendinger and Glotfelty, 1990
					3.30	21 °C, gas stripping	Friesen et al., 1993
					4.94	gas stripping	Alaee et al., 1996
fluoranthene	0.00123	0.00129	0.957		0.65	20 °C, gas stripping	ten Hulscher et al., 1992
pyrene	0.0006	0.00065	0.920	1.21 ± 0.23	1.89	gas stripping	Southworth, 1979
					1.10	gas stripping	Mackay and Shiu, 1981
benzo[*b*]fluoranthene		0.0006			0.051	20 °C, gas stripping	ten Hulscher et al., 1992
benzo[*k*]fluoranthene	5.2×10^{-8}	3.2×10^{-6}	0.016		0.044	20 °C, gas stripping	ten Hulscher et al., 1992
benzo[*a*]pyrene	7.0×10^{-7}	1.51×10^{-5}	0.046		0.034	20 °C, gas stripping	ten Hulscher et al., 1992
benzo[*ghi*]perylene	2.25×10^{-5} [c]	9.7×10^{-7}	0.075		0.027	20 °C, gas stripping	ten Hulscher et al., 1992
indeno[1,2,3-*cd*]pyrene					0.029	20 °C, gas stripping	ten Hulscher et al., 1992
benzene	12700	22.79	557		562	gas stripping	Mackay et al., 1979
					554	concn ratio	Leighton and Calo, 1981
					746	20 °C, EPICS	Yurteri et al., 1987
					588	gas stripping	Ashworth et al., 1988
					535	EPICS	Ashworth et al., 1988
					535	static head space method	Robbins et al., 1993
					570	equilibrium head space	Ettre et al., 1993
					649	29 °C, EPICS	Hansen et al., 1993
					481	EPICS	Dewulf et al., 1993
					550	gas stripping	Alaee et al., 1996
chlorobenzene	1580	4.30	367	315 ± 5.6	382	gas stripping	Mackay et al., 1979
					330	concn ratio	Leighton and Calo, 1981
					314	gas stripping	Mackay and Shiu, 1981
					398	gas stripping	Warner et al., 1987
					319	20 °C, EPICS	Yurteri et al., 1987
					346	20 °C, EPICS	Ashworth et al., 1988
					324	equilibrium head space	Ettre et al., 1993
1,2-dichlorobenzene	196	0.803	244.2	195 ± 4.5	193	gas stripping	Mackay and Shiu, 1981
					122	20 °C, gas stripping	Oliver, 1985
					197	gas stripping	Warner et al., 1987
					159	20 °C, EPICS	Ashworth et al., 1988
1,3-dichlorobenzene	307	0.816	376.1		182	20 °C, gas stripping	Oliver, 1985
					267	gas stripping	Warner et al., 1987
					290	EPICS	Ashworth et al., 1988
1,4-dichlorobenzene	90.2	0.565	159.8	244 ± 5.6	330	concn ratio	Leighton and Calo, 1981
					240	gas stripping	Mackay and Shiu, 1981
					152	20 °C, gas stripping	Oliver, 1985
					276	gas stripping	Warner et al., 1987
					190	20 °C, EPICS	Yurteri et al., 1987
					365	EPICS	Ashworth et al., 1988
1,2,3-trichlorobenzene	28	0.116	242.0	127 ± 2.1	127	gas stripping	Mackay and Shiu, 1981
					90	gas stripping	Oliver, 1985
					72	20 °C, gas stripping	ten Hulscher et al., 1992
1,2,4-trichlorobenzene	61	0.220	276.7		122	gas stripping	Oliver, 1985
					144	gas stripping	Warner et al., 1987
					101	gas stripping	ten Hulscher et al., 1992
1,3,5-trichlorobenzene	32	0.0292	1096		193	gas stripping	Oliver, 1985
					192	gas stripping	ten Hulscher et al., 1992
1,2,3,4-tetrachlorobenzene	5.2	0.0361	143.9		69.9	gas stripping	Oliver, 1985
					62	gas stripping	ten Hulscher et al., 1992
1,2,3,5-tetrachlorobenzene	9.8	0.0167	587.7	160 ± 2.8	159	gas stripping	Mackay and Shiu, 1981
					99	20 °C, gas stripping	ten Hulscher et al., 1992
1,2,4,5-tetrachlorobenzene	0.72	0.00588	122.4		101	gas stripping	Oliver, 1985
pentachlorobenzene	0.22	0.00260	84.72		71.9	gas stripping	Oliver, 1985

Table 1 (Continued)

compound	vap pres P^S/Pa[a]	solubility C^S/mol·m^{-3} [a]	H_{calc}/ Pa·m^3·mol^{-1} P^S/C^S	H_{exptl}/ Pa·m^3·mol^{-1} (this work)	H(lit.)/ Pa·m^3· mol^{-1}	method	ref
hexachlorobenzene	0.0023	1.76 × 10^{-5}	131.0		59.0	gas stripping	ten Hulscher et al., 1992
					48.6	gas stripping	Oliver, 1985
					172	gas stripping	Warner et al., 1987
					26243	26 °C, EPICS, see text	Hansen et al., 1993
					41	gas stripping	ten Hulscher et al., 1992
1-chloronaphthalene		0.138		36.3 ± 1.0			
2-chloronaphthalene		0.0718		33.5 ± 1.60			
bromobenzene	552	2.61	211	250 ± 7.3	247	gas stripping	Mackay and Shiu, 1981
					256	30 °C, EPICS	Hansen et al., 1993
2-methyl-1-propanol	1500	1092	1.37	2.73 ± 0.12			
1-heptanol	24	14.97	1.603	5.62 ± 0.4			
2-pentanone	4720	691	6.83	8.47 ± 0.64			
2-heptanone	500	37.7	13.3	17.1 ± 0.38			
acetophenone	45	45.77	0.982	1.08 ± 0.048	0.921	gas stripping	Betterton, 1991

[a] Selected values from Mackay et al. (1992a,b, 1995). [b] EPICS, equilibrium partitioning in closed system. [c] Supercooled liquid value.

TLV-45 or TLV-70 temperature baths. Nitrogen was passed through a Matheson Model-70 low-pressure line regulator, the gas flow rate was monitored by a Brook's Sho-Rate 150 Model 1355-02B-V rotameter, and the exit gas flow rate was measured by a soap bubble flow meter. The concentration of the dissolved chemicals in water was determined by gas chromatography or fluorescence.

Analytical Methods. Fluorescence spectrophotometry was used for the analysis of polynuclear aromatic hydrocarbons because of its high sensitivity. Gas chromatographic analysis followed by a purge-and-trap analysis was used for the volatile halogenated hydrocarbons and the more hydrophilic compounds. Although the stripping efficiency was low for the alcohols and ketones even at an elevated temperature and long stripping times (Kuo et al., 1977), it was possible to monitor the concentration change using a fixed purge time.

The fluorescence measurements were performed on an Aminco-Bowman spectrophotofluorometer (American Instrument Ltd.). Excitation and emission wavelengths were chosen on the basis of the compound's spectra. The fluorescent intensity was measured in a quartz cell containing 1−2 mL of sample directly taken from the stripping apparatus or cyclohexane extract of the water sample and recorded by a Hewlett-Packard XY recorder Model 7740.

The gas chromatograph was a Hewlett-Packard GC Model 5840 equipped with both a dual flame ionization detector, an electron capture detector, and a Model 7675A purge-and-trap sampler. The analytical column was a 50 m long, 0.5 mm o.d. borosilicate glass open tubular column coated with SE 30. The carrier gas and purge gas were both nitrogen or helium. The chemical was purged from the water sample into a Tenax-GC trap in the sample and then thermosorbed, and the sorbed chemical was swept directly onto the analytical column by the carrier gas. Peak area was integrated by a Hewlett-Packard GC Terminal Model 5840A.

Experimental Procedure. Aqueous hydrocarbon solutions were prepared by slowly stirring an excess amount of the chemical in a 1 L Erlenmeyer flask containing doubly distilled water for 24 h. A quantity of the aqueous solution was drawn off from the flask and added to the stripping vessel usually diluted with water to obtain an optimal concentration. After the desired experimental temperature and gas flow rate were stablized, samples were taken for analysis. Determinations were done for each chemical individually, not as mixtures.

Experimental difficulties were encountered for compounds with a Henry's law constant less than 10 Pa·m^3·mol^{-1} (such as pyrene), in which only small concentration changes were obtained, the error in the slope can be appreciable, and the accuracy of determination is reduced. For the hydrophilic compounds, the stripping rate was very low even at elevated temperature, and long stripping times were required.

Results and Discussion

Table 1 gives the experimental results and previously reported values for comparison together with the ratio P^S/C^S. Results of triplicate measurements or more (up to 15 for naphthalene) of Henry's law constants are tabulated with their standard deviations in Table 1. In most cases agreement is satisfactory, i.e., within ±8%, since there is a tendency for H or K_{AW} to be fairly constant or change systematically within a homologous series (despite large differences in P^S and C^S). Some reported data for compounds which were not the subject of experimental determination are included to facilitate examination of the dependence of H on molecular structure.

The polycyclic aromatic hydrocarbons show a trend of diminishing H with increasing molecular mass from about 44 Pa·m^3·mol^{-1} for naphthalene to nearly 1 Pa·m^3·mol^{-1} for pyrene to values of approximately 0.03 for the larger multiring compounds. The corresponding values of K_{AW} are a factor of RT or 2479 lower at 25 °C. The chlorobenzenes range similarly from 560 Pa·m^3·mol^{-1} for benzene to nearly 100 Pa·m^3·mol^{-1} for penta- and hexachlorobenzenes. As molecular mass increases, the vapor pressure falls more rapidly than the solubility. The alcohols and ketones have low H values in the range 1 to 18 Pa·m^3·mol^{-1}.

In some cases, there are considerable differences between reported values of H and disagreement with estimates from P^S/C^S. Notable in this respect are the experimental values of 2-methylnaphthalene and hexachlorobenzene reported by Hansen et al. (1993) who used the EPICS method which was developed for volatile organic solutes. A case can be made for determination of P^S, C^S, and H since this allows for a consistency check of the data.

Conclusion

Experimental data have been reported from the Henry's law constants of selected aromatic and polycyclic aromatic hydrocarbons, alcohols, and ketones at 25 °C.

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