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Density and Surface Tension of Aqueous H_2SO_4 at Low Temperature

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The density and surface tension of 0.12 to 0.75 acid mass fraction in aqueous sulfuric acid solutions have been measured in the temperature region from (220 to 300) K. The density has been expressed as a polynomial of the acid mass fraction and temperature, fitting the new measurements together with the tabulated values from *International Critical Tables* (1928). Older literature data for the surface tension of aqueous sulfuric acid has been corrected for systematic errors and used with the new data to derive a parametrization of the surface tension as a function of acid mass fraction and temperature.

1. Introduction

Sulfuric acid aerosols are found throughout the lower and middle stratosphere (e.g., Turco et al., 1982; Hoffmann, 1990; Dye et al., 1992; Yue et al., 1994). The composition of the binary system water sulfuric acid may be calculated from the thermodynamic state functions of aqueous sulfuric acid and those of water vapor (Gmitro and Vermeulen, 1961; Zeleznik, 1991; Clegg and Brimblecombe, 1995; Massucci et al., 1996). The actual aerosol composition depends on the local water vapor pressure and temperature, but in addition also upon the density, the surface tension, and the particle size through the Kelvin equation given by Renninger et al. (1981)

$$\ln\left(\frac{P_r}{P_\infty}\right) = \frac{2M_{H_2O}\gamma}{RTr\rho}\left(1+\frac{w}{\rho}\frac{\partial\rho}{\partial w}\right) \qquad (1)$$

where r is the radius of curvature, P_∞ is the water vapor pressure over a plane surface, and P_r is the water vapor pressure over a curved surface with radius r. M_{H_2O} is the molar mass of water, γ the surface tension, T the temperature, w the mass fraction sulfuric acid, and ρ is the density.

During our laboratory studies of the physical properties of the stratospheric sulfuric acid aerosols, it became evident that no measurements of the density and the surface tension have been published for temperatures below 273 K. In previous theoretical studies of the sulfuric acid aerosol composition and the Kelvin effect, values for the density and surface tension at low temperatures were by necessity extrapolated from data obtained above 273 K (Doyle, 1961; Nair and Vohra, 1975; Steele and Hamill, 1981, Renninger et al., 1981). The surface tension is also a crucial parameter in the microphysical description of nucleation and homogeneous freezing of sulfate aerosols (Jensen et al., 1991; MacKenzie et al., 1995; Tabazadeh et al., 1997a,b).

The density measurements on sulfuric acid were reviewed and summarized in *International Critical Tables* (1928); with the exception of a graphical display of the measured density for a solution of 0.36 mass fraction sulfuric acid from 300 K to ca. 220 K (Garrett et al., 1948; Garrett and Woodruff, 1951), no data have appeared since. Several studies of the surface tension have been published, but so far all of the data have not been compared. Different

Table 1. Density at Mass Fractions *w* of (1 − *w*) Water + *w* Sulfuric Acid

T/K	ρ/kg m^{-3}	T/K	ρ/kg m^{-3}
$w = 0.123$			
300	1079.47	276	1089.88
298	1079.86	273	1090.85
297	1080.22	270	1091.77
292	1082.51	269	1092.60
288	1084.16	265	1094.07
284	1085.93	262	1094.57
280	1087.78		
$w = 0.291$			
303	1204.67	263	1232.67
299	1207.32	263	1232.67
294	1211.23	259	1235.90
288	1214.93	250	1242.09
281	1219.84	247	1243.64
270	1227.56	243	1246.01
268	1229.24	241	1246.99
$w = 0.503$			
301	1391.84	256	1403.67
297	1395.45	246	1438.64
291	1401.95	243	1442.26
284	1406.99	236	1447.33
278	1411.47	229	1452.69
269	1418.86	221	1460.25
262	1425.00		
$w = 0.585$			
302	1473.08	262	1508.49
298	1476.54	252	1517.57
292	1482.20	248	1521.08
286	1487.73	246	1522.96
281	1492.34	241	1528.29
275	1497.20	235	1533.45
269	1502.62		
$w = 0.672$			
303	1569.25	254	1615.08
297	1574.85	248	1619.41
294	1578.04	245	1623.96
290	1581.40	235	1633.93
283	1586.66	230	1639.15
277	1593.49	228	1641.98
265	1604.76	221	1649.67
263	1605.51		
$w = 0.765$			
306	1675.52	261	1719.20
298	1682.20	250	1731.50
297	1682.53	249	1731.85
291	1689.20	243	1738.94
283	1696.60	242	1739.68
277	1702.43	233	1749.83
271	1708.39		

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Table 2. Coefficients in a Polynomial Expansion of the Density of Aqueous Sulfuric Acid

$$\rho(w,\ T)/\mathrm{kg{\cdot}m^{-3}} = \sum_{i=0}^{10}\sum_{j=0}^{4}\rho_{i,j}w^i(T/\mathrm{K} - 273.15)^j$$

$\rho_{i,j}$	$j = 0$	1	2	3	4
$i = 0$	999.8426	334.5402×10^{-4}	-569.1304×10^{-5}		
1	547.2659	-530.0445×10^{-2}	118.7671×10^{-4}	599.0008×10^{-6}	
2	526.2950×10^{1}	372.0445×10^{-1}	120.1909×10^{-3}	-414.8594×10^{-5}	119.7973×10^{-7}
3	-621.3958×10^{2}	-287.7670	-406.4638×10^{-3}	111.9488×10^{-4}	360.7768×10^{-7}
4	409.0293×10^{3}	127.0854×10^{1}	326.9710×10^{-3}	-137.7435×10^{-4}	-263.3585×10^{-7}
5	-159.6989×10^{4}	-306.2836×10^{1}	136.6499×10^{-3}	637.3031×10^{-5}	
6	385.7411×10^{4}	408.3714×10^{1}	-192.7785×10^{-3}		
7	-580.8064×10^{4}	-284.4401×10^{1}			
8	530.1976×10^{4}	809.1053			
9	-268.2616×10^{4}				
10	576.4288×10^{3}				

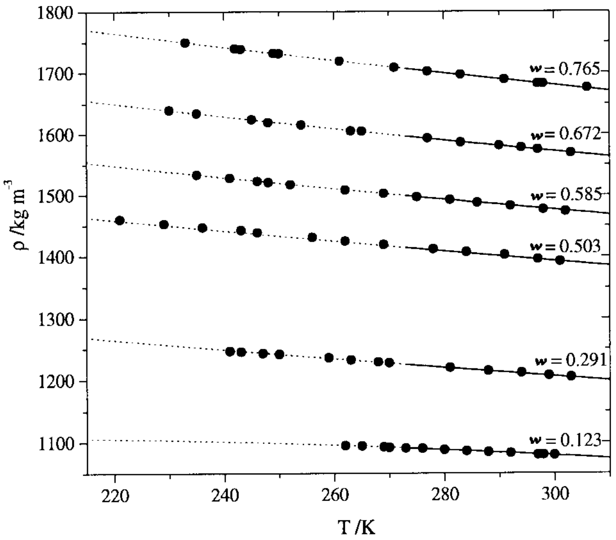

Figure 1. Map of new measurements of the density of aqueous sulfuric acid. Filled circles, data from this work; solid curves, interpolated values from *International Critical Tables* (1928); dotted curves, extrapolated values from *International Critical Tables* (1928). The numbers on the curves are the mass fractions of the solutions.

methods have been employed in the previous studies of the surface tension: Morgan and Davis (1916) used the drop-weight method; Sabinina and Terpugow (1935) and Hoffmann and Seemann (1960) obtained their data by the capillary-height method; Suggitt et al. (1949) used the differential maximum bubble pressure technique.

2. Experimental Section

The sulfuric acid solutions were prepared from concentrated sulfuric acid of p.a. quality (Fluka) and ion-exchanged, distilled water. The composition of the solutions was subsequently determined by titration before and after use. The relative uncertainty of the acid mass fraction in the individual solutions is better than ±0.2%.

The densities were determined by weighing a small, solid quartz bulb immersed in the acid. The volume of the quartz bulb and its temperature dependence were calibrated against doubly distilled, degassed water. The temperature was controlled to within ±0.5 K by circulating a coolant through a double-walled beaker containing the acid. The whole setup was sealed from the moist atmosphere in a nitrogen flushed glovebag. Each data point represents the average of at least three individual measurements, and the derived densities are believed to be accurate to ±1 kg $\mathrm{m^{-3}}$ (3δ).

Table 3. Surface Tension at Mass Fractions *w* of (1 − *w*) Water + *w* Sulfuric Acid

T/K	γ/J $\mathrm{m^{-2}}$	δ^a/J $\mathrm{m^{-2}}$	T/K	γ/J $\mathrm{m^{-2}}$	δ^a/J $\mathrm{m^{-2}}$
		$w = 0.123$			
295	0.073 04	0.000 08	276	0.075 72	0.000 20
293	0.073 33	0.000 08	272	0.076 25	0.000 12
288	0.074 11	0.000 32	270	0.076 60	0.000 24
286	0.074 44	0.000 30	268	0.077 09	0.000 33
281	0.075 15	0.000 15	265	0.077 36	0.000 17
		$w = 0.291$			
294	0.075 15	0.000 12	258	0.077 48	0.000 13
288	0.075 74	0.000 15	253	0.077 78	0.000 17
286	0.076 27	0.000 14	250	0.078 03	0.000 25
278	0.076 50	0.000 09	248	0.078 19	0.000 55
273	0.076 80	0.000 09	246	0.077 81	0.000 37
268	0.077 00	0.000 20	243	0.078 19	0.000 18
263	0.077 22	0.000 12			
		$w = 0.503$			
297	0.076 16	0.000 07	278	0.077 39	0.000 28
296	0.076 28	0.000 10	271	0.078 00	0.000 29
293	0.076 46	0.000 08	261	0.078 81	0.000 16
291	0.076 53	0.000 26	251	0.079 19	0.000 24
285	0.077 01	0.000 19	243	0.080 34	0.000 15
283	0.077 24	0.000 09	241	0.080 90	0.000 13
		$w = 0.585$			
297	0.075 86	0.000 03	266	0.077 42	0.000 08
293	0.076 13	0.000 03	263	0.077 57	0.000 06
288	0.076 23	0.000 03	258	0.078 10	0.000 13
283	0.076 54	0.000 10	253	0.078 36	0.000 22
278	0.076 77	0.000 07	250	0.078 45	0.000 16
273	0.077 01	0.000 05	243	0.078 79	0.000 12
		$w = 0.672$			
297	0.074 86	0.000 11	266	0.076 01	0.000 11
293	0.075 00	0.000 20	258	0.076 37	0.000 12
286	0.075 31	0.000 09	251	0.076 60	0.000 15
283	0.075 40	0.000 15	243	0.077 05	0.000 16
278	0.075 58	0.000 09	238	0.077 20	0.000 36
273	0.075 87	0.000 11	233	0.077 41	0.000 36

[a] δ is the standard deviation.

The surface tension was derived from a computer image analysis of a sessile bubble trapped in the aqueous acid (Hansen and Rødsrud, 1991; Hansen, 1993). The temperature control and the environment were identical to that described for the density measurements. Each data point represents the average of at least 20 individual measurements. The precision of the instrument was determined by measurements of pure water as a function of temperature between each series of measurements of H_2SO_4. On the basis of this, the accuracy is believed to be better than ±0.0005 J $\mathrm{m^{-2}}$.

3. Results and Comparison of Data

(a) Density. The measured densities are presented in Table 1; they agree with interpolated literature values in

Table 4. Corrections to the Surface Tension at Mass Fractions w of $(1 - w)$ Water + w Sulfuric Acid Determined by Morgan and Davis (1916)

	$T = 273$ K		$T = 303$ K		$T = 323$ K	
w	γ/J m^{-2}	correction/J m^{-2}	γ/J m^{-2}	correction/J m^{-2}	γ/J m^{-2}	correction/J m^{-2}
0.0000	0.075 75	−0.000 11	0.071 03	0.000 16	0.067 59	0.000 32
0.0467	0.075 35	0.000 07	0.071 15	0.000 29	0.067 96	0.000 42
0.0893	0.075 27	0.000 22	0.071 31	0.000 39	0.068 26	0.000 51
0.1640	0.075 66	0.000 43	0.071 87	0.000 57	0.068 95	0.000 66
0.2273	0.076 07	0.000 60	0.072 62	0.000 70	0.069 80	0.000 78
0.2818	0.076 56	0.000 74	0.073 10	0.000 82	0.070 52	0.000 89
0.3290	0.076 91	0.000 86	0.073 60	0.000 93	0.071 24	0.000 98
0.3705	0.077 02	0.000 97	0.073 97	0.001 02	0.071 70	0.001 06
0.4071	0.077 05	0.001 07	0.074 27	0.001 10	0.072 12	0.001 14
0.4238	0.077 11	0.001 11	0.074 34	0.001 14	0.072 35	0.001 17
0.4397	0.077 02	0.001 16	0.074 39	0.001 18	0.072 38	0.001 21
0.4546	0.076 97	0.001 20	0.074 42	0.001 22	0.072 44	0.001 24
0.4688	0.076 80	0.001 24	0.074 46	0.001 25	0.072 51	0.001 28
0.4806			0.074 37	0.001 29		
0.4823	0.076 57	0.001 28	0.074 34	0.001 29	0.072 50	0.001 31
0.4951	0.076 41	0.001 32	0.074 30	0.001 33	0.072 49	0.001 34
0.5410	0.075 82	0.001 46	0.074 06	0.001 45	0.072 41	0.001 45
0.5786	0.075 17	0.001 57	0.073 80	0.001 54	0.072 24	0.001 54
0.6108	0.074 45	0.001 66	0.073 29	0.001 63	0.072 01	0.001 62
0.6623	0.073 39	0.001 79	0.072 47	0.001 75	0.071 34	0.001 74
0.7018	0.072 21	0.001 88	0.071 63	0.001 84	0.070 41	0.001 83
0.7295	0.071 49	0.001 94	0.071 00	0.001 90	0.069 85	0.001 88
0.7584	0.070 63	0.001 99	0.070 03	0.001 95	0.068 87	0.001 93
0.7969	0.069 06	0.002 05	0.068 66	0.002 01	0.067 24	0.001 99
0.8306	0.067 22	0.002 08	0.066 72	0.002 04	0.065 47	0.002 01
0.8626			0.064 12	0.002 04		
0.8729	0.064 12	0.002 07	0.063 29	0.002 04	0.062 20	0.002 01
0.8929	0.062 06	0.002 05	0.061 24	0.002 02	0.060 10	0.001 98
0.9182			0.058 34	0.001 96		
0.9262			0.057 53	0.001 94	0.056 70	0.001 92
0.9363	0.057 06	0.001 94	0.055 56	0.001 89	0.056 42	0.001 91
0.9449	0.056 18	0.001 91	0.055 44	0.001 89	0.054 67	0.001 86
0.9515			0.054 68	0.001 86		
0.9566	0.054 98	0.001 88	0.054 28	0.001 85	0.053 61	0.001 83
0.9612	0.054 44	0.001 86	0.053 67	0.001 83	0.053 08	0.001 81
0.9689			0.052 79	0.001 80		
0.9732			0.052 33	0.001 78		
0.9920			0.050 23	0.001 71		
0.9946			0.050 08	0.001 70		
0.9980			0.049 63	0.001 68	0.049 06	0.001 66
1.0000			0.049 62	0.001 68		

Table 5. Coefficients in a Polynomial Expansion of the Surface Tension of Aqueous Sulfuric Acid

$$\gamma(w,\ T)/\mathrm{J{\cdot}m^{-2}} = \sum_{i=0}^{7}\sum_{j=0}^{3}\gamma_{i,j}w^{i}(T/\mathrm{K} - 273.15)^{j}$$

$\gamma_{i,j}$	$j = 0$	1	2	3
$i = 0$	75.640×10^{-3}	-13.818×10^{-5}	-31.807×10^{-8}	
1	23.033×10^{-4}	-35.768×10^{-4}	26.948×10^{-5}	-38.605×10^{-7}
2	-12.361×10^{-2}	55.742×10^{-3}	-39.071×10^{-4}	55.691×10^{-6}
3	12.516×10^{-1}	-30.759×10^{-2}	21.360×10^{-3}	-30.419×10^{-5}
4	-42.193×10^{-1}	83.258×10^{-2}	-57.881×10^{-3}	82.448×10^{-5}
5	64.995×10^{-1}	-11.910×10^{-1}	83.310×10^{-3}	-11.865×10^{-4}
6	-47.546×10^{-1}	86.407×10^{-2}	-60.908×10^{-3}	86.653×10^{-5}
7	13.185×10^{-1}	-24.979×10^{-2}	17.746×10^{-6}	-25.204×10^{-5}

the temperature region above 273 K (*International Critical Tables*, 1928). Figure 1 shows our new measurements together with the interpolated and extrapolated values from *International Critical Tables* (1928). The data in *International Critical Tables* (1928) are interpolated values and presented for all integral values of the mass percent from 1 to 100 and for the temperatures (0, 10, 15, 20, 25, 30, 40, 50, 60, 80, and 100) °C.

The sulfuric acid density is often required in different numerical algorithms. We therefore present a parametrization of the density as a polynomial function of acid content and temperature using our new data and the tabulated values (*International Critical Tables*, 1928) for the $\{w, T/\mathrm{K}|\ 0.10{-}0.90;\ 273{-}323\}$ region

$$\rho(w,\ T)/\mathrm{kg{\cdot}m^{-3}} = \sum_{i=0}^{10}\sum_{j=0}^{4}\rho_{i,j}w^{i}(T/\mathrm{K} - 273.15)^{j} \qquad (2)$$

where $\rho(w, T)$ is the density, w is the mass fraction sulfuric acid, and T is the temperature. $\rho_{\mathrm{o,o}}$ was taken as the density of standard mean ocean water at 0 °C, and the $\rho_{\mathrm{o},j}$ coefficients were determined by fitting a polynomial to the tabulated densities of standard mean ocean water (Marsh, 1987). A quadratic polynomial is sufficient to fit the density of water from 0 to 40 °C with a maximum error of less than 0.15 kg m^{-3} and a root-mean-square (rms) error of less than 0.08 kg m^{-3}. Similarly, the coefficients $\rho_{i,\mathrm{o}}$ were obtained from a polynomial fit to the densities of aqueous sulfuric acid at 0 °C (*International Critical Tables*, 1928).

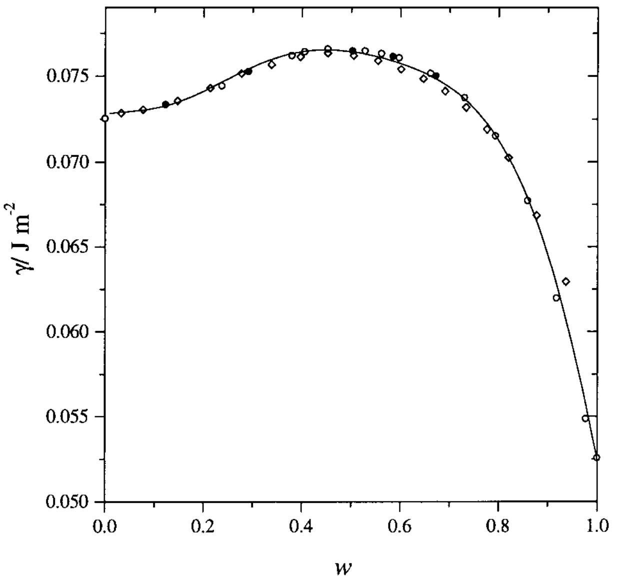

Figure 2. Comparison of surface tension data for aqueous sulfuric acid at 293 K. Open circles, data from Sabinina and Terpugow (1935); diamonds, Hoffmann and Seemann (1960); filled circles, data or interpolated data from this work The solid curve shows the modeled surface tension.

In this case a polynomial of degree 10 is needed to model the 0 °C data such that the largest deviation is less than ± 0.4 kg m^{-3} (rms error 0.12 kg m^{-3}). The $\rho_{i,0}$ and $\rho_{0,j}$ terms were subsequently constrained to the above-mentioned values during the global fit. The final parametrization containing 32 terms, shown in Table 2, is continuous and well-behaved in the entire mass fraction−temperature region $\{w, T/\mathrm{K}|0.10-0.90; 210-323\}$ and reproduces the tabulated values from *International Critical Tables* (1928) within ± 0.5 kg m^{-3}, as well as 95% of our new density data to better than ± 1 kg m^{-3}.

(b) Surface Tension. The new measurements of the surface tension are summarized in Table 3. To compare the available data, some of the older results need to be corrected.

Correction of Data. The data of Morgan and Davis (1916) obtained by the drop-weight method relies on an experimental relationship between the surface tension, γ, the weight of the drop, w, and the inner radius of the dropping tip, r

$$\gamma = \frac{w}{2\pi r} \cdot \phi\left(\frac{r}{\sqrt[3]{v}}\right) \tag{3}$$

where ϕ is an experimentally determined function of the ratio between tip radius, r, and the cube root of the drop volume, v. Morgan and Davis (1916) assumed that $\phi/2\pi r$ was constant, and they derived its value ($2\pi r/g\phi = 9.36 \times 10^{-5}$ s^{-2}) from experiments assuming known surface tension values of pure water. Later, Harkins and Brown (1919) carried out extensive measurements to determine the function ϕ. Suggitt et al. (1949) made a limited reanalysis of the Morgan and Davis data using improved values for the surface tension of water and assuming ϕ to be a linear function in the relevant region of $r/v^{1/3}$. A closer inspection of the Morgan and Davis (1916) and the Harkins and Brown (1919) data shows that ϕ is far from being linear in the interval of interest, $r/v^{1/3} = 0.6221$ to 0.8751, and is best approximated by a quadratic function

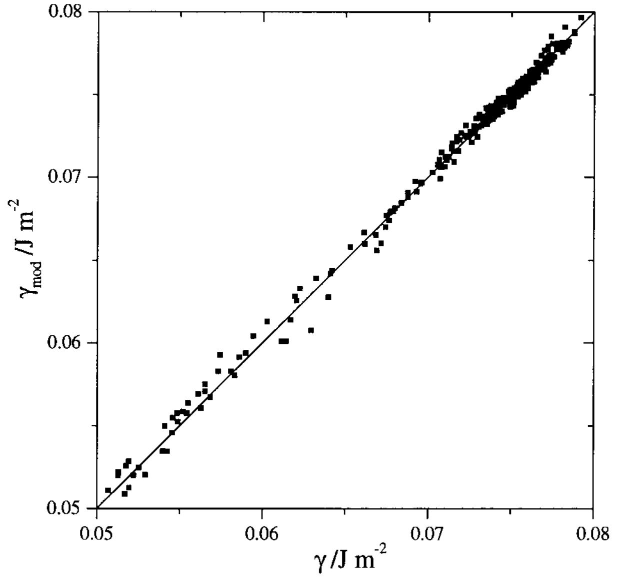

Figure 3. Comparison between observed and modeled surface tension/J m^{-2} of aqueous sulfuric acid.

$$\frac{g}{2\pi} \cdot \phi\left(\frac{r}{\sqrt[3]{v}}\right) = 1.4004 + 2.8159 \cdot \left(\frac{r}{\sqrt[3]{v}}\right) - 1.6460 \cdot \left(\frac{r}{\sqrt[3]{v}}\right)^2 \tag{4}$$

where the acceleration of free fall, g, was taken as 9.81 m s^{-2} as in the original paper by Harkins and Brown (1919). The radius of the tip used in the Morgan and Davis data was found from their water measurements by solving eqs 3 and 4 using the new, recommended values for the surface tension of water at (0, 30, and 50) °C (Vargaftik et al., 1983). We found the same lack of thermal variation in the tip radius as Suggitt et al. (1949), but slightly larger values: $r = 0.2691$ cm (0 °C), 0.2691 cm (30 °C), and 0.2690 cm (50 °C). The corrections of the surface tensions of the Morgan and Davis data for aqueous sulfuric acid, presented together with the original data in Table 4, were then obtained from eqs 3 and 4 using this tip dimension.

Sabinina and Terpugow (1935) obtained their data at (10, 20 30, 40, and 50) °C by the method of capillary rise. They have given their concentrations in mole percent sulfuric acid. Their measured results for pure water are generally slightly lower than today's recommended values (Vargaftik et al., 1983): 0.074 01 J m^{-2} vs 0.074 23 (10 °C), 0.072 53 J m^{-2} vs 0.072 75 (20 °C), 0.071 03 J m^{-2} vs 0.071 20 (30 °C), 0.069 54 J m^{-2} vs 0.069 60 (40 °C) and 0.067 80 J m^{-2} vs 0.067 94 (50 °C). On the average, their values for water are 0.2% lower than today's recommendation. This may arise from an error in the radius of the capillary. On the other hand, they do not describe the procedure for how they determined the concentrations or give the source of their air and sulfuric acid densities. Thus, it is not possible to validate or correct their data.

The data of Suggitt et al. (1949) were obtained by the differential maximum bubble pressure technique at 25 °C assuming that the surface tension of water at the same temperature was 0.071 97 J m^{-2}. The recommended value today is 0.071 99 J m^{-2} (Vargaftik et al., 1983) so that the correction is far less than the estimated experimental uncertainty.

Hoffmann and Seemann (1960) measured the surface tension of aqueous sulfuric acid at 15 °C and 20 °C by the capillary height method. They specify the acid concentration by the density of the solution, and, according to the number of digits, the uncertainty in concentration is ca. $\pm 0.5\%$. We interpolated the values from *International*

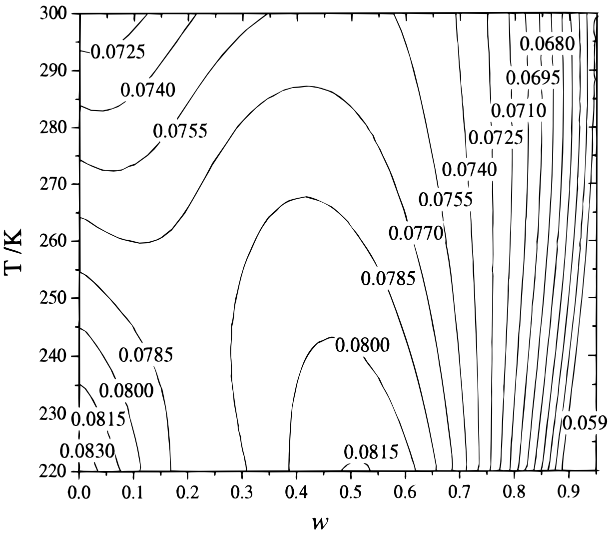

Figure 4. Calculated surface tension/J m^{-2} in all the relevant (w, T/K) space: {0.0−0.95, 220−300}.

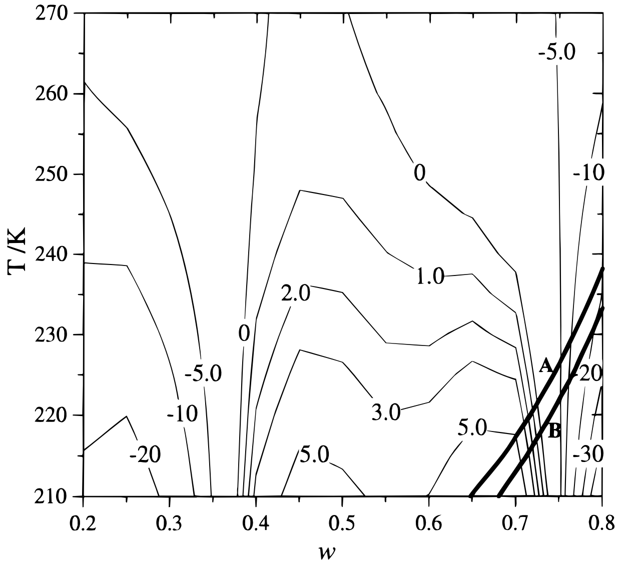

Figure 5. Difference between the sulfuric acid surface tension calculated using eq 5 and the parametrization of Tabazadeh et al. (1997b). The compositions of the stratospheric sulfuric acid aerosol (Tabazadeh et al., 1997c) are given for mid-latitude conditions by curve A (∼16 km, 100 hPa, 5 ppm water) and curve B (∼20 km, 60 hPa, 4.5 ppm water).

Critical Tables (1928) to find the corresponding mass fractions of the solutions.

After these corrections, all the different data sets appeared to be in fair agreement with our new data. Figure 2 shows a comparison of the literature data and our data at $T = 293$ K. The modeled surface tension, see below, is also included.

Parametrization. The complete set of data was fitted to a polynomial function of the form

$$\gamma(w, T)/\mathrm{J\cdot m^{-2}} = \sum_{i=0}^{7}\sum_{j=0}^{3}\gamma_{i,j}w^{i}(T/\mathrm{K} - 273.15)^{j} \qquad (5)$$

where $\gamma(w, T)$ is the surface tension, w is the mass fraction of sulfuric acid, and T is the temperature.

The coefficients $\gamma_{i,\mathrm{o}}$ were determined from the corrected $T = 273$ K data of Morgan and Davis (1916). A polynomial of order 7 in the mass fraction is needed to fit the data within 0.7 mJ m^{-2} (rms error 0.25 mJ m^{-2}). The $\gamma_{\mathrm{o},j}$ coefficients from pure water (Vargaftik et al., 1983) and a third-order polynomial in T are needed to fit the 0 to 50 °C data to better than 0.01 mJ m^{-2}. These coefficients were then constrained during the global fit. The parameters of the final fit (rms error 0.5 mJ m^{-2}) are collected in Table 5, while Figure 3 shows a comparison between the observed and modeled surface tensions. The modeled surface tension is presented in Figure 4; the expression is well-behaved in all the relevant (w, T) space.

The difference between the above and a recent parametrization of the surface tension (Tabazadeh et al., 1997b), based upon the data of Sabinina and Terpugow (1935), is shown in Figure 5. There are only minor differences between the two parametrizations in the region {w, T/K; 0.2−0.75| 220−270}. Figure 5 also includes composition lines of sulfuric acid aerosols of stratospheric compositions at mid-latitudes, ∼16 km (curve A) and ∼20 km (curve B), calculated from the parametrization presented by Tabazadeh et al. (1997c). As can be seen, the deviation for these conditions is significant.

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