

| T(K) ± 0.01 | wt% NH_4HSO_4 ± 0.03 | Solids present |
|--------------------|---------------------------------------------|-------------------|
| 253.05 | 34.45 | ice |
| 259.05 | 29.58 | ice |
| 263.05 | 24.15 | ice |
| 264.85 | 21.99 | ice |
| 269.15 | 12.96 | ice |
| 271.15 | 5.66 | ice |

Table 1. $\text{NH}_4\text{HSO}_4(\text{aq})$ composition in equilibrium with ice at several temperatures.

| SO_4^{2-} (m) | H^+ (m) | NH_4^+ (m) | T (K) ± 0.01 | $a_{\text{H}_2\text{O}(s)}$ | uncertainty $a_{\text{H}_2\text{O}(s)}$ | $\Sigma\sigma$ (m) | $m_{\text{H}^+}/m_{\text{SO}_4^{2-}}$ |
|------------------------|------------------|---------------------|---------------------|-----------------------------|--------------------------------------------|--------------------|---------------------------------------|
| 0.98 \pm 0.05 | 0.96 \pm 0.03 | 1.00 \pm 0.05 | 273.00 | 0.993 | ± 0.015 | 0.00 \pm 0.08 | 0.98 \pm 0.06 |
| | | | 278.15 | 0.936 | 0.015 | | |
| | | | 283.19 | 0.889 | 0.010 | | |
| | | | 288.23 | 0.846 | 0.007 | | |
| | | | 293.21 | 0.799 | 0.005 | | |
| | | | 298.25 | 0.762 | 0.005 | | |
| 2.51 \pm 0.08 | 2.44 \pm 0.05 | 2.49 \pm 0.08 | 268.14 | 0.969 | 0.020 | -0.09 \pm 0.10 | 0.97 \pm 0.03 |
| | | | 273.09 | 0.932 | 0.015 | | |
| | | | 278.26 | 0.867 | 0.015 | | |
| | | | 283.30 | 0.838 | 0.010 | | |
| | | | 288.28 | 0.800 | 0.007 | | |
| | | | 293.21 | 0.764 | 0.005 | | |
| 298.13 | 0.743 | 0.005 | | | | | |
| 3.57 \pm 0.09 | 3.53 \pm 0.06 | 3.54 \pm 0.09 | 259.07 | 0.916 | 0.035 | -0.07 \pm 0.10 | 1.00 \pm 0.02 |
| | | | 264.05 | 0.864 | 0.035 | | |
| | | | 269.13 | 0.828 | 0.020 | | |
| | | | 274.02 | 0.798 | 0.015 | | |
| | | | 279.20 | 0.767 | 0.010 | | |
| | | | 284.08 | 0.735 | 0.007 | | |
| | | | 289.06 | 0.702 | 0.005 | | |
| | | | 294.19 | 0.672 | 0.005 | | |
| 299.13 | 0.642 | 0.005 | | | | | |
| 6.14 \pm 0.10 | 6.11 \pm 0.07 | 6.11 \pm 0.10 | 253.22 | 0.983 | 0.040 | -0.06 \pm 0.20 | 1.00 \pm 0.01 |
| | | | 258.12 | 0.951 | 0.035 | | |
| | | | 263.08 | 0.878 | 0.035 | | |
| | | | 268.12 | 0.832 | 0.020 | | |
| | | | 273.14 | 0.796 | 0.020 | | |
| | | | 278.09 | 0.758 | 0.015 | | |
| | | | 283.18 | 0.722 | 0.015 | | |
| | | | 288.28 | 0.689 | 0.007 | | |
| | | | 293.21 | 0.658 | 0.005 | | |
| 298.31 | 0.628 | 0.005 | | | | | |

Table 2. Ice activity, $a_{\text{H}_2\text{O}(s)}$, data for several $\text{NH}_4\text{HSO}_4(\text{aq})$ compositions. Concentrations of SO_4^{2-} , H^+ , and NH_4^+ are in molality. The sum of the charges, $\Sigma\sigma$, and the ratio, $m_{\text{H}^+}/m_{\text{SO}_4^{2-}}$, are provided as a check for self-consistency in the measured data.

| SO_4^{2-} (m) | H^+ (m) | NH_4^+ (m) | T (K) ± 0.01 | $a_{\text{H}_2\text{O}(s)}$ | uncertainty $a_{\text{H}_2\text{O}(s)}$ | $\Sigma\sigma$ (m) | $m_{\text{H}^+}/m_{\text{SO}_4^{2-}}$ |
|------------------------|------------------|---------------------|---------------------|-----------------------------|--------------------------------------------|--------------------|---------------------------------------|
| 8.93 \pm 0.20 | 8.82 \pm 0.09 | 9.05 \pm 0.20 | 243.06 | 0.693 | ± 0.040 | +0.01 \pm 0.30 | 0.99 \pm 0.01 |
| | | | 248.01 | 0.659 | 0.040 | | |
| | | | 253.02 | 0.612 | 0.035 | | |
| | | | 258.08 | 0.574 | 0.030 | | |
| | | | 263.13 | 0.534 | 0.030 | | |
| | | | 268.03 | 0.506 | 0.020 | | |
| | | | 273.18 | 0.484 | 0.015 | | |
| | | | 278.20 | 0.459 | 0.010 | | |
| | | | 283.19 | 0.439 | 0.007 | | |
| | | | 288.23 | 0.417 | 0.005 | | |
| | | | 293.27 | 0.396 | 0.005 | | |
| 12.72 \pm 0.20 | 12.45 \pm 0.10 | 12.63 \pm 0.20 | 263.10 | 0.445 | 0.040 | -0.36 \pm 0.30 | 0.98 \pm 0.01 |
| | | | 268.15 | 0.410 | 0.040 | | |
| | | | 273.21 | 0.386 | 0.035 | | |
| | | | 278.21 | 0.370 | 0.030 | | |
| | | | 283.19 | 0.354 | 0.020 | | |
| | | | 288.18 | 0.338 | 0.010 | | |
| | | | 293.22 | 0.324 | 0.007 | | |
| | | | 298.31 | 0.309 | 0.005 | | |
| 19.97 \pm 0.30 | 19.58 \pm 0.20 | 19.64 \pm 0.30 | 283.23 | 0.634 | 0.030 | -0.72 \pm 0.50 | 0.98 \pm 0.01 |
| | | | 288.17 | 0.324 | 0.020 | | |
| | | | 293.33 | 0.161 | 0.010 | | |
| | | | 298.22 | 0.083 | 0.007 | | |

Table 2 (cont). Ice activity, $a_{\text{H}_2\text{O}(s)}$, data for several $\text{NH}_4\text{HSO}_4(\text{aq})$ compositions.

Concentrations of SO_4^{2-} , H^+ , and NH_4^+ are in molality. The sum of the charges, $\Sigma\sigma$, and the ratio, $m_{\text{H}^+}/m_{\text{SO}_4^{2-}}$, are provided as a check for self-consistency in the measured data.

| wt% NH ₄ HSO ₄ ±0.03 | T(K) ±0.01 | wt% H ⁺ ±0.003 | Solids present |
|-----------------------------------------------|---------------|------------------------------|-------------------|
| 47.0 | 254.90* | 0.408 | |
| | 250.95 | 0.410 | letovicite |
| | 249.15 | 0.411 | letovicite |
| | 247.15 | 0.411 | letovicite |
| | 245.15 | 0.413 | letovicite |
| 49.2 | 259.42* | 0.427 | |
| | 259.05 | 0.429 | letovicite |
| | 257.15 | 0.432 | letovicite |
| | 255.15 | 0.436 | letovicite |
| | 253.05 | 0.443 | letovicite |
| 55.0 | 272.22* | 0.482 | |
| | 257.05 | 0.492 | letovicite |
| | 255.15 | 0.493 | letovicite |
| | 253.15 | 0.494 | letovicite |
| | 251.15 | 0.496 | letovicite |
| 60.4 | 281.94* | 0.525 | |
| | 277.15 | 0.532 | letovicite |
| | 275.15 | 0.538 | letovicite |
| | 273.05 | 0.539 | letovicite |
| | 271.15 | 0.541 | letovicite |

Table 3. H⁺ concentration of NH₄HSO₄(aq) solutions in equilibrium with letovicite, (NH₄)₃H(SO₄)₂(s), at several temperatures. *These temperatures correspond to the initial H⁺ concentration of the aqueous solution when letovicite first precipitates, as determined by the extrapolation method shown in Figure 5.