Table S1: Summary of selected prior work characterizing size-resolved organic acids and methanesulfonate (MSA).

| Location | Selected Species | Reference | | | |
|--|--|---|--|--|--|
| Arctic | | | | | |
| Arctic | MSA, oxalate, succinate | Kerminen et al. (1999) | | | |
| Asia | | | | | |
| Chengdu, China | Oxalate, succinate, adipate, maleate, phthalate | Cheng et al. (2015) Huang et al. (2019) | | | |
| Shanghai, China | MSA, oxalate | Ding et al. (2017) | | | |
| North, west, and south of Japan | Oxalate, succinate | Gao et al. (2003) | | | |
| Gwangju, Korea | Oxalate | Park and Kim (2014) | | | |
| Baoji, China Mt. Tai, China Okinawa Island, Japan | Succinate, phthalate | Wang et al. (2017) | | | |
| Xi'an, China | Oxalate, succinate, adipate, maleate, phthalate | Wang et al. (2012) | | | |
| Hong Kong | Oxalate, succinate | Yao et al. (2002) | | | |
| Europe | | | | | |
| Finokalia, Greece | MSA, oxalate, succinate | Bardouki et al. (2003) | | | |
| Sagres, Portugal Melpitz, Germany | MSA, oxalate, succinate, adipate | Neusüss et al. (2000) | | | |
| Melpitz, Germany Falkenberg, Germany Goldlauter, Germany Leipzig, Germany Dresden, Germany | Oxalate, succinate | van Pinxteren et al. (2014) | | | |
| Po Valley, Italy | Oxalate | Sandrini et al. (2016) | | | |
| North America | | | | | |
| Central California | MSA, succinate, maleate, oxalate | Maudlin et al. (2015) | | | |
| California | Oxalate | Murphy et al. (2009) | | | |
| Hayden and Winkelman, Arizona | Oxalate | Sorooshian et al. (2012) | | | |
| Thompson Farm and Shoals, New England | Oxalate, succinate, maleate, MSA | Ziemba et al. (2011) | | | |
| South America | | | | | |
| Rondônia, Brazil | MSA, oxalate, succinate, maleate, adipate, phthalate | Blazso et al. (2003) Decesari et al. (2006) Falkovich et al. (2005) | | | |
| Other | | | | | |
| North Pacific East Asia | Oxalate, succinate | Mochida et al. (2003) | | | |
| Tudor Hill, Bermuda | Oxalate | Turekian et al. (2003) | | | |

Table S2: MOUDI sample set details including sample start date and time, sampling flow rate, sample duration, and average values of both the temperature of the MOUDI housing and relative humidity (RH) of ambient air. Sets marked with "*" are sets with gravimetric data.

| Sample ID | Start Date (Local) | Flow (L min ⁻¹) | Run Time (h) | Temp. (°C) | RH (%) | Sample ID | Start Date (Local) | Flow (L min ⁻¹) | Run Time (h) | Temp. (°C) | RH (%) |
|--------------|--------------------------|-----------------------------------|--------------------|---------------|-----------|---------------|--------------------------|-----------------------------------|--------------------|---------------|-----------|
| MO1 | 07/19/18 12:40 | 29.6 | 24 | 30.5 | 59.0 | MO31* | 02/13/19 15:33 | 29.9 | 49 | 35.8 | 65.7 |
| MO2 | 07/23/18 11:29 | 29.6 | 54 | 31.7 | 66.8 | MO33 | 02/23/19 5:00 | 29.8 | 48 | 34.3 | 58.1 |
| MO4* | 07/25/18 19:16 | 30.3 | 119 | 34.4 | 69.0 | MO34 | 03/04/19 14:05 | 29.4 | 48 | 35.3 | 57.9 |
| MO5 | 07/30/18 19:17 | 28.8 | 42 | 33.5 | 66.7 | MO36* | 03/12/19 15:50 | 29.3 | 48 | 39.9 | 56.8 |
| MO6 | 08/06/18 14:33 | 27.1 | 48 | 34.6 | 63.3 | MO37 | 03/20/19 | 30.0 | 48 | 38.8 | 55.1 |
| MO7 | 08/14/18 | 27.9 | 48 | 34.9 | 78.3 | MO38 | 03/30/19 | 29.6 | 48 | 36.4 | 54.0 |
| MO8 | 08/22/18 13:46 | 29.0 | 48 | 35.7 | 78.2 | MO40 * | 04/08/19 14:30 | 29.6 | 48 | 41.4 | 57.6 |
| MO9 | 09/01/18 5:00 | 27.5 | 48 | 34.9 | 68.4 | MO41 | 04/16/19 14:30 | 29.1 | 48 | 38.7 | 57.7 |
| MO10 | 09/10/18 14:42 | 29.0 | 48 | 36.7 | 65.2 | MO42 | 04/24/19 14:30 | 29.1 | 48 | 40.3 | 53.7 |
| M011 | 09/18/18 14:12 | 27.1 | 48 | 35.8 | 68.3 | MO44* | 05/04/19 5:00 | 28.6 | 48 | 37.0 | 59.8 |
| MO12 | 09/26/18 13:53 | 27.5 | 48 | 37.0 | 70.9 | MO45 | 05/13/19 14:30 | 28.7 | 48 | 37.3 | 61.8 |
| MO14* | 10/06/18 5:00 | 26.1 | 48 | 32.0 | 73.1 | MO46 | 05/21/19 14:30 | 28.7 | 48 | 39.0 | 72.2 |
| M015 | 10/15/18 14:37 | 29.7 | 48 | 37.3 | 67.6 | MO47 | 05/29/19 14:24 | 28.9 | 48 | 39.3 | 64.5 |
| MO16 | 10/23/18 14:18 | 29.2 | 48 | 37.6 | 67.7 | MO48* | 06/08/19 5:00 | 28.0 | 48 | 38.9 | 62.6 |
| MO17 | 11/06/18 15:42 | 30.0 | 48 | 36.5 | 60.6 | MO50 | 06/17/19 14:20 | 28.8 | 48 | 39.2 | 64.4 |
| MO18 | 11/10/18 5:00 | 29.5 | 48 | 36.7 | 61.9 | MO51 | 06/25/19 14:18 | 27.8 | 50 | 36.2 | 77.1 |
| MO19 | 11/19/18 13:55 | 31.4 | 48 | 35.8 | 61.4 | MO53* | 07/03/19 14:43 | 26.9 | 48 | 38.8 | 60.9 |
| MO20 | 11/27/18 14:16 | 30.2 | 48 | 34.8 | 60.8 | MO54 | 07/13/19 5:00 | 28.8 | 48 | 36.8 | 66.4 |
| MO21 | 12/05/18 13:57 | 30.5 | 48 | 34.8 | 72.0 | MO55 | 07/22/19 14:40 | 28.8 | 48 | 38.0 | 75.4 |
| MO22 | 12/15/18 5:00 | 29.6 | 48 | 32.7 | 78.5 | MO56 | 07/30/19 14:37 | 26.7 | 48 | 35.0 | 76.1 |
| MO23 | 12/24/18 13:30 | 26.4 | 48 | 29.7 | 81.8 | MO57* | 08/07/19 14:18 | 27.5 | 48 | 33.0 | 94.1 |
| MO24 | 12/31/18 14:45 | 30.2 | 48 | 35.8 | 84.6 | MO59 | 08/17/19 5:00 | 28.2 | 48 | 37.8 | 85.9 |
| MO26 | 01/01/19 13:30 | 24.1 | 48 | 35.0 | 77.2 | MO60 | 08/26/19 14:31 | 28.2 | 48 | 37.3 | 92.7 |
| MO27 | 01/09/19 14:48 | 23.9 | 48 | 36.2 | 65.3 | MO61 | 09/03/19 14:28 | 29.4 | 48 | 36.3 | 62.1 |
| MO28 | 01/19/19 5:00 | 25.0 | 48 | 33.1 | 63.5 | MO62* | 09/11/19 14:34 | 27.8 | 48 | 36.5 | 77.0 |
| MO29 | 01/28/19 14:17 | 29.5 | 48 | 34.5 | 63.3 | MO64 | 09/21/19 5:00 | 27.0 | 48 | 37.5 | 67.2 |
| MO30 | 02/05/19 13:30 | 29.8 | 48 | 34.4 | 60.7 | MO65* | 09/30/19 14:30 | 27.2 | 48 | 38.4 | 65.3 |

| Table S3: Water-soluble species analyzed with their respective recovery \pm standard deviation |
|---|
| (SD) and limit of detection (LOD) in aqueous concentration units quantified using ICP-QQQ |
| (elements: Al to V) and IC (ions: adipate to sulfate). LOD units for elements and ions are ppt and |
| ppb, respectively. |

| Species | Recovery ± SD (%) | LOD | | |
|-----------|----------------------|--------|--|--|
| Al | 96 ± 7 | 29.474 | | |
| As | 98 ± 10 | 7.945 | | |
| Cd | 102 ± 11 | 4.194 | | |
| Κ | 93 ± 18 | 10.48 | | |
| Ni | 97 ± 8 | 2.837 | | |
| Pb | 99 ± 8 | 0.503 | | |
| Ti | 101 ± 10 | 39.046 | | |
| V | 95 ± 9 | 1.353 | | |
| Adipate | 101 ± 4 | 22.655 | | |
| Chloride | 103 ± 7 | 2.144 | | |
| Maleate | 100 ± 3 | 6.97 | | |
| MSA | 102 ± 6 | 12.316 | | |
| Nitrate | 106 ± 12 | 8.917 | | |
| Oxalate | 100 ± 2 | 12.312 | | |
| Phthalate | 99 ± 2 | 20.685 | | |
| Sodium | 104 ± 8 | 43.476 | | |
| Succinate | 98 ± 9 | 11.046 | | |
| Sulfate | 101 ± 3 | 11.982 | | |

| Factors | 5 |
|--------------------------------------|---------|
| QRobust | 3075 |
| QTrue | 4369 |
| QRob/Exp | 0.41 |
| Q/Q _{Exp} > 6 | - |
| DISP %dQ | -0.0004 |
| DISP swaps | 0 |
| Factors with BS mapping < 100% | 1 |
| BS-DISP % cases with swaps | 0 |

Table S4: Summary of positive matrix factorization (PMF) and error estimation diagnostics.

Table S5: Percentage that each organic acid and MSA contributes to the total mass for all $(0.056 - 18 \ \mu\text{m})$, submicrometer, and supermicrometer size ranges, as well as for each individual stage. These percentages were calculated as the mean based on the 11 simultaneously collected MOUDI sets for gravimetric and chemical analysis. "Unresolved" represents the percent difference between the total mass and the following measured constituents: black carbon (BC), and water-soluble ions (Na⁺, NH₄⁺, Mg²⁺, Ca²⁺, dimethylamine (DMA), trimethylamine (TMA), and diethylamine (DEA), methanesulfonate (MSA), pyruvate, adipate, succinate, maleate, oxalate, phthalate, Cl⁻, NO₃⁻, and SO₄²⁻) and elements (Ag, Al, As, Ba, Cd, Co, Cr, Cs, Cu, Fe, Hf, K, Mn, Mo, Nb, Ni, Pb, Rb, Se, Sn, Sr, Ti, Tl, V, Y, Zn, and Zr). Error associated with BC is reported as the standard deviation in measured BC as a function of size. Cells marked with "-" denote values of zero.

| Stages | Total | Phthalate | Adipate | Succinate | Maleate | Oxalate | MSA | Unresolved | BC Error |
|--------------|-------|-----------|---------|-----------|---------|---------|-------|------------|-----------------|
| All | 0.80% | 0.03% | 0.03% | 0.03% | 0.04% | 0.64% | 0.02% | 33.74% | 21.72% |
| < 1 µm | 0.78% | 0.03% | 0.01% | 0.04% | 0.06% | 0.60% | 0.03% | 17.78% | 34.27% |
| > 1 µm | 0.84% | 0.04% | 0.06% | 0.02% | - | 0.71% | 0.01% | 69.10% | 3.59% |
| 10 - 18 | 0.63% | - | 0.04% | 0.04% | - | 0.56% | _ | 79.43% | 26.47% |
| 5.6 - 10 | 0.70% | - | 0.21% | 0.02% | - | 0.45% | 0.02% | 76.76% | 1.04% |
| 3.2 – 5.6 | 0.56% | 0.03% | 0.06% | 0.02% | - | 0.44% | - | 73.36% | 2.95% |
| 1.8 - 3.2 | 0.98% | 0.07% | 0.04% | 0.03% | - | 0.84% | _ | 67.60% | 3.48% |
| 1.0 - 1.8 | 1.01% | 0.03% | 0.03% | 0.01% | 0.01% | 0.93% | 0.01% | 63.21% | 8.68% |
| 0.56 - 1.0 | 1.06% | 0.03% | 0.01% | 0.05% | 0.13% | 0.80% | 0.04% | 36.58% | 15.25% |
| 0.32 - 0.56 | 0.88% | 0.04% | 0.01% | 0.07% | 0.06% | 0.65% | 0.04% | 16.04% | 29.51% |
| 0.18 - 0.32 | 0.55% | 0.03% | 0.01% | 0.01% | 0.01% | 0.46% | 0.03% | 2.99% | 43.55% |
| 0.10 - 0.18 | 0.56% | - | 0.04% | 0.05% | - | 0.45% | 0.02% | 7.97% | 85.41% |
| 0.056 - 0.10 | 0.49% | - | - | 0.03% | - | 0.46% | _ | 63.88% | 49.91% |

- 1 Table S6: Pearson's correlation matrix (r values) of water-soluble species across all MOUDI
- 2 sizes $(0.056 18 \,\mu\text{m})$. Blank boxes indicate p-values exceeding 0.05 and thus deemed to be
- 3 statistically insignificant. Similar correlation matrices can be seen for submicrometer and
- 5 phthalate.

| Al | 1.00 | | | | | | | | | | | | | | | | | | |
|------------|------|------|------|------|------|------|------|------|------|------|------|------|------------|------|------|------|------|------|------|
| Ti | 0.49 | 1.00 | | _ | | | | | | | | | | | | | | | |
| K | 0.57 | | 1.00 | | _ | | | | | | | | | | | | | | |
| Rb | 0.63 | | 0.48 | 1.00 | | _ | | | | | | | | | | | | | |
| V | | | | 0.36 | 1.00 | | _ | | | | | | | | | | | | |
| Ni | 0.35 | 0.34 | | 0.41 | 0.79 | 1.00 | | | | | | | | | | | | | |
| As | | 0.41 | | | | | 1.00 | | | | | | | | | | | | |
| Cd | | 0.28 | | | 0.67 | 0.69 | | 1.00 | | _ | | | | | | | | | |
| Pb | 0.44 | 0.33 | 0.30 | 0.29 | 0.32 | 0.42 | | 0.44 | 1.00 | | | | | | | | | | |
| Na | 0.36 | 0.38 | | | | | | | | 1.00 | | | | | | | | | |
| Cl | 0.54 | 0.36 | 0.41 | | | | | | | 0.78 | 1.00 | | _ | | | | | | |
| NO3 | 0.49 | | 0.44 | 0.31 | | | | | | 0.56 | 0.49 | 1.00 | | | | | | | |
| SO4 | | | 0.32 | 0.46 | 0.44 | | | | | | | 0.50 | 1.00 | | _ | | | | |
| MSA | | | | 0.37 | | | | | | 0.42 | | 0.35 | 0.59 | 1.00 | | | | | |
| Ad | | | | | | | | | | | | | | | 1.00 | | | | |
| Su | 0.36 | | | 0.67 | | | | | | 0.38 | | | 0.43 | 0.67 | | 1.00 | | | |
| Ma | | | | | | | | | | | | | | | 0.45 | | 1.00 | | _ |
| Ox | 0.42 | | | 0.71 | 0.44 | 0.45 | | | | | | 0.31 | 0.69 | 0.53 | | 0.76 | | 1.00 | |
| Ph | 0.41 | 0.36 | | 0.48 | | | | | | 0.53 | 0.35 | 0.31 | 0.34 | 0.58 | 0.43 | 0.73 | | 0.63 | 1.00 |
| | Al | Ti | K | Rb | V | Ni | As | Cd | Pb | Na | Cl | NO3 | SO4 | MSA | Ad | Su | Ma | Ox | Ph |

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| Gradian | | Pe | eak Bins (µr | n) | Peak Values (ng m ⁻³) | | | | | |
|-----------|------------------------------------|-----------------------|----------------------------------|---|-----------------------------------|--------------------|-------------|--------------------|-----------------------------|-------------------|
| Species | All | SWM18 | Trans | NEM | SWM19 | All | SWM18 | Trans | NEM | SWM19 |
| Phthalate | 0.32-0.56 1.8-3.2 | 0.32-0.56 3.2-5.6 | 0.32-0.56 3.2-5.6 | 0.18-0.32 3.2-5.6 | 0.56-1.0 1.8-3.2 | 2.1 1.2 | 5.7 0.81 | 9.2 2.9 | 0.68 1.5 | 1.4 1.8 |
| Adipate | 0.10-0.18 0.32-0.56 5.6-10.0 | 0.32-0.56 5.6-10.0 | 0.32-0.56 1.0-1.8 5.6-10.0 | 0.10-0.18 1.8-3.2 5.6-10.0 | 0.10-0.18 0.56-1.0 3.2-5.6 | 1.1 1.0 0.95 | 1.7 0.93 | 2.2 1.4 0.86 | 0.86 0.37 0.75 | 1.6 1.4 2.3 |
| Succinate | 0.32-0.56 1.8-3.2 | 0.32-0.56 1.8-3.2 | 0.18-0.32 0.56-1.0 3.2-5.6 | 0.10-0.18 0.32-0.56 1.8-3.2 5.6-10.0 | 0.56-1.0 1.8-3.2 | 3.1 0.69 | 7.8 1.1 | 1.5 4.0 0.26 | 0.52 2.0 0.73 0.26 | 2.5 0.41 |
| Maleate | 0.56-1.0 | 0.56-1.0 | 0.56-1.0 | 0.32-0.56 | 0.56-1.0 | 5.1 | 9.9 | 1.7 | 0.94 | 11 |
| Oxalate | 0.32-0.56 1.8-3.2 | 0.32-0.56 1.8-3.2 | 0.32-0.56 1.8-3.2 | 0.32-0.56 1.8-3.2 | 0.56-1.0 1.8-3.2 | 32 20 | 35 26 | 58 30 | 31 18 | 28 18 |
| MSA | 0.32-0.56 5.6-10.0 | 0.32-0.56 | 0.32-0.56 1.8-3.2 | 0.32-0.56 5.6-10.0 | 0.32-0.56 5.6-10.0 | 2.0 0.043 | 3.5 | 2.2 0.15 | 1.2 0.034 | 2.3 0.11 |

Table S7: Size distribution peaks for each season and respective concentrations. "All" represents the cumulative dataset.



Figure S1: Size-resolved comparison of (black curve) total mass and (colored curve) the sum of measured organic acids and MSA for the (a) 2018 southwest monsoon (SWM18), (b) transitional period (Trans), (c) northeast monsoon (NEM), and (d) 2019 southwest monsoon (SWM19) seasons. Solid lines are the averages and shaded areas represent one standard deviation. The average percent contribution of the organic acids and MSA to total mass is provided for each size bin. These plots are based on data from the 11 MOUDI chemical sets with accompanying gravimetric measurements. It should be noted that SWM18 and Trans each only one gravimetric set and thus standard deviations are unavailable.



Figure S2: Cumulative size distributions for each organic acid and MSA. The transparent lines are the individual sets, while the thick colored lines and transparent shaded areas represent the mean and standard deviation, respectively, of the cumulative sets.



Figure S3: WCWT maps of phthalate for (a) 2018 southwest monsoon (SWM18), (b) transitional period (Trans), (c) northeast monsoon (NEM), and (d) 2019 southwest monsoon (SWM19) for all sizes.



Figure S4: Same as Figure S3 except for adipate.



Figure S5: Same as Figure S3 except for succinate.



Figure S6: Same as Figure S3 except for maleate.



Figure S7: Same as Figure S3 except for oxalate.



Figure S8: Same as Figure S3 except for MSA.

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