**SUPPORTING INFORMATION FOR**

**Impact of Dry Intrusion Events on Composition and Mixing State of Particles During Winter ACE-ENA Study**

Jay M. Tomlin1, Kevin A. Jankowski1, Daniel P. Veghte3,4, Swarup China4, Peiwen Wang5,   
Matthew Fraund6, Johannes Weis6, Guangjie Zheng7,8, Yang Wang8,9, Felipe Rivera-Adorno1,   
Shira Raveh-Rubin10, Daniel A. Knopf5, Jian Wang7,8, Mary K. Gilles6, Ryan C. Moffet11,   
Alexander Laskin1,2\*

1Department of Chemistry, 2Department of Earth Atmospheric and Planetary Sciences, Purdue University, West Lafayette, IN 47907, USA  
3Center for Electron Microscopy and Analysis, Ohio State University, Columbus, OH 43212, USA  
4Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory, Richland, WA 99354, USA  
5School of Marine and Atmospheric Sciences, Stony Brook University, Stony Brook, NY 11794, USA  
6Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA  
7Center for Aerosol Science and Engineering, Department of Energy, Environmental and Chemical Engineering, Washington University in St. Louis, St. Louis,MO 63130, USA 8Environmental and Climate Science Department, Brookhaven National Laboratory, Upton, NY 11973, USA 9Department of Civil, Architectural and Environmental Engineering, Missouri University of Science and Technology, Rolla, MO 65409, USA  
10Department of Earth and Planetary Sciences, Weizmann Institute of Science, Rehovot 76100, Israel  
11Sonoma Technology, Inc., Petaluma, CA 94954, USA

*Correspondence to*: Alexander Laskin(alaskin@purdue.edu)

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| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Summary of analyzed G1 aircraft samples | | | | | | | | | |
| **Sample date (YYYY-MM-DD)** | **TRAC hole no.** | **Sampling time (hh:mm:ss) (UTC)** | **Sampling altitude MSL (avg ± stdev.) (m)** | **Boundary layer height (m)** | **Dry intrusion event** | **Aerosol particle concentration (PCASP) (avg ± stdev.) (#/cm3)** | **Wind speed**  **(avg ± stdev.) (m/s)** | **Analysis** | **No. of particles analyzed** |
| 2018-01-19 | 33 | 14:14:55-14:24:56 | 288±351 (BL) | 1000 | No | 76.3±47.9 | 6.1±1.7 | CCSEM | 842 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-01-21 | 33 | 9:52:20-9:59:33 | 2673±4 (FT) | 1600 | No | 15.8±4.0 | 11.9±2.6 | CCSEM | 1160 |
| 2018-01-21 | 36 | 10:13:29-10:20:32 | 153±4 (BL) | 1600 | No | 42.3±6.7 | 11.7±0.6 | CCSEM | 1193 |
| 2018-01-21 | 46 | 11:23:54-11:30:57 | 1832±6 (FT) | 1600 | No | 11.2±3.6 | 15.6±0.4 | CCSEM | 409 |
| 2018-01-21 | 48 | 11:38:00-11:45:03 | 40±6 (BL) | 1600 | No | 33.9±7.6 | 12.7±0.5 | CCSEM | 1171 |
| 2018-01-21 | 53 | 12:13:15-12:20:18 | 1226±16 (BL) | 1600 | No | 97.6±111.0 | 14.2±1.5 | STXM | 377 |
| 2018-01-21 | 58 | 12:48:27-12:55:30 | 1818±4 (FT) | 1600 | No | 14.7±3.7 | 16.7±0.6 | STXM | 806 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-01-24 | 42 | 13:31:46-13:38:49 | 227±145 (BL) | 2100 | Yes | 32.7±6.0 | 7.0±0.6 | CCSEM & STXM | 1713 & 15 |
| 2018-01-24 | 49 | 14:21:07-14:28:10 | 1678±26 (BL) | 2100 | Yes | 34.9±86.0 | 6.6±0.9 | STXM | 68 |
| 2018-01-24 | 51 | 14:35:14-14:42:17 | 2175±3 (FT) | 2100 | Yes | 23.4±9.1 | 15.1±0.8 | CCSEM & STXM | 2991 & 28 |
| 2018-01-24 | 59 | 15:31:36-15:38:39 | 1029±5 (BL) | 2100 | Yes | 28.4±27.3 | 6.0±0.7 | STXM | 283 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-01-25 | 26 | 12:07:08-12:14:11 | 1366±10 (BL) | 1650 | Yes | 55.2±26.7 | 2.9±1.2 | CCSEM | 2402 |
| 2018-01-25 | 28 | 12:21:13-12:28:16 | 1566±47 (BL) | 1650 | Yes | 176.9±137.9 | 3.8±1.2 | STXM | 561 |
| 2018-01-25 | 42 | 13:59:51-14:06:53 | 1889±136 (FT) | 1650 | Yes | 24.0±37.2 | 3.3±1.0 | STXM | 23 |
| 2018-01-25 | 44 | 14:13:56-14:20:59 | 879±560 (BL) | 1650 | Yes | 61.2±26.6 | 3.3±1.1 | STXM | 166 |
| 2018-01-25 | 46 | 14:28:01-14:35:03 | 236±11 (BL) | 1650 | Yes | 35.5±6.9 | 3.6±0.7 | STXM | 97 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-01-26 | 39 | 11:49:55-11:56:58 | 1130±22 (BL) | 1200 | Yes | 54±27.7 | 3.7±0.8 | CCSEM | 1216 |
| 2018-01-26 | 49 | 13:00:22-13:07:25 | 1408±394 (FT) | 1200 | Yes | 63.9±91.4 | 4.4±0.5 | STXM | 467 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-01-28 | 48 | 12:52:48-12:59:51 | 1866±2 (FT) | 1700 | No | 18.1±5.1 | 8.9±0.5 | STXM | 398 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-01-30 | 33 | 9:42:52-9:49:55 | 2485±160 (FT) | 1650 | No | 23.8±9.6 | 6.8±0.9 | CCSEM | 3386 |
| 2018-01-30 | 44 | 11:00:25-11:07:28 | 1259±38 (BL) | 1650 | No | 73.6±87.4 | 4.4±1.8 | STXM | 284 |
| 2018-01-30 | 64 | 13:21:21-13:28:24 | 1508±2 (BL) | 1650 | No | 10.5±3.2 | 4.6±0.4 | CCSEM & STXM | 955 & 268 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-02-01 | 47 | 12:11:29-12:18:32 | 787±137 (BL) | 850 | No | 61.3±23.1 | 6.1±1.6 | CCSEM | 967 |
| 2018-02-01 | 58 | 13:29:02-13:36:04 | 43±8 (BL) | 850 | No | 86.4±22.3 | 3.8±1.1 | STXM | 76 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-02-08 | 47 | 14:53:14-15:00:16 | 157±10 (BL) | 2150 | No | 73.8±10.0 | 9.3±0.8 | CCSEM | 949 |
| 2018-02-08 | 51 | 15:21:23-15:28:26 | 1479±3 (BL) | 2150 | No | 31.2±6.5 | 8.1±0.7 | CCSEM | 912 |
| 2018-02-08 | 57 | 16:03:41-16:10:44 | 2347±23 (FT) | 2150 | No | 17.8±6.8 | 11.6±0.4 | CCSEM | 1305 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-02-11 | 66 | 14:16:06-14:23:09 | 1390±5 (BL) | 1950 | No | 60.5±54.1 | 5.7±0.8 | CCSEM | 1053 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-02-15 | 34 | 13:10:09-13:17:12 | 2450±7 (FT) | 2150 | Yes | 13.3±3.8 | 10.0±0.9 | CCSEM | 1383 |
| 2018-02-15 | 42 | 14:06:33-14:13:36 | 1162±6 (BL) | 2150 | Yes | 131.1±167.7 | 1.8±1.1 | CCSEM | 1022 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-02-16 | 41 | 13:23:46-13:30:49 | 168±188 (BL) | 2100 | Yes | 23.7±4.7 | 2.3±0.5 | CCSEM | 1524 |
| 2018-02-16 | 43 | 13:37:51-13:44:54 | 1057±3 (BL) | 2100 | Yes | 26.0±9.6 | 4.4±0.9 | CCSEM & STXM | 1536 & 5 |
| 2018-02-16 | 48 | 14:13:05-14:20:08 | 2181±19 (FT) | 2100 | Yes | 142.8±17.2 | 4.3±0.4 | CCSEM & STXM | 1331 & 10 |
| 2018-02-16 | 51 | 14:34:14-14:41:17 | 1888±4 (BL) | 2100 | Yes | 200.5±522.8 | 1.6±0.9 | STXM | 34 |
| 2018-02-16 | 58 | 15:23:34-15:30:37 | 2193±5 (FT) | 2100 | Yes | 131.1±26.8 | 3.2±0.6 | CCSEM & STXM | 1681 & 179 |
| 2018-02-16 | 66 | 16:19:58-16:24:01 | 1744±20 (BL) | 2100 | Yes | 31.7±6.2 | 2.1±0.9 | CCSEM | 1362 |
|  |  |  |  |  |  |  |  |  |  |
| 2018-02-19 | 37 | 11:33:44-11:40:47 | 43±4 (BL) | 1800 | Yes | 16.4±4.4 | 7.8±0.8 | CCSEM & STXM | 3429 & 11 |
| 2018-02-19 | 52 | 13:19:24-13:26:29 | 4045±14 (FT) | 1800 | Yes | 37.2±8.2 | 4.1±1.9 | CCSEM & STXM | 1278 & 21 |
| 2018-02-19 | 54 | 13:33:32-13:40:35 | 4057±8 (FT) | 1800 | Yes | 35.7±7.3 | 3.2±1.3 | STXM | 73 |
| 2018-02-19 | 58 | 14:01:44-14:08:47 | 4057±9 (FT) | 1800 | Yes | 35.8±7.3 | 3.2±1.3 | STXM | 82 |
|  | | | | | | | | | |

**Table S1. Samples analyzed for the study by CCSEM and STXM/NEXAFS and parameter discussed (sample date, sampling time, average sampling altitude, average sampling altitude, boundary layer height, synoptic condition, average aerosol particle concentration, wind speed near sea level, and number of particles analyzed by each type of analysis).**

**Graphical user interface, chart, map

Description automatically generatedFigure S1. Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) (Stein et al., 2015; Rolph et al., 2017) back trajectories for dry intrusion periods utilizing global data assimilation system (GDAS1) achieved model data at three starting** **elevations 100 m (red), 2000 m (blue), 3000 m (green). A) 2018-01-24; B) 2018-01-25; C) 2018-01-26; D) 2018-02-15; E) 2018-02-16; F) 2018-02-19.**

**Diagram

Description automatically generatedFigure S2. HYSPLIT back trajectories for each sampling day during non-dry intrusion periods utilizing GDAS1 achieved model data at three starting elevation 100 m (red), 2000 m (blue), 3000 m (green). A) 2018-01-19; B) 2018-01-21; C) 2018-01-28; D) 2018-01-30; E) 2018-02-01; F) 2018-02-08; G) 2018-02-11.**

**Chart

Description automatically generatedFigure S3. Flight summaries of altitude and particle concentration vs. time during dry intrusion periods: A) 2018-01-24; B) 2018-01-25; C) 2018-01-26; D) 2018-02-15; E) 2018-02-16; F) 2018-02-19. Blue shaded regions show time periods of particle sampling.**

**A picture containing chart

Description automatically generatedFigure S4. Flight summaries of altitude and particle concentration vs. time during non-dry intrusion periods: A) 2018-01-19; B) 2018-01-21; C) 2018-01-28; D) 2018-01-30; E) 2018-02-01; F) 2018-02-08; G) 2018-02-11. Blue shaded regions show time periods of particle sampling.**

# CCSEM-EDX particle classification schemes

**Chart, bar chart

Description automatically generated**All analyzed particles were combined before grouping into clusters using a *k*-means clustering algorithm (Hartigan and Wong, 1979) and utilizing the quantitative elemental contribution of 15 selected elements (e.g. C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Mn, Fe, and Cu) acquired by the CCSEM/EDX analysis with Cu excluded in the analysis due to the material of the particle substrate. The square root of elemental contribution of individual particles were implemented in the *k*-means clustering to allow for the larger weighting of trace elements (Rebotier and Prather, 2007). The initial condition of the *k*-means algorithm is defined with eight initial centroids (i.e., cluster centers) for the entire data set. We then combined clusters with similar mean elemental contributions and narrowed the classification into four major clusters: (1) Mixed Sea Salt, (2) Aged Sea Salt, (3) Ammonium Nitrate/Sulfates, (4) Carbonaceous, as shown below.

**Figure S5. Mean elemental contribution (logarithmic scale) for each particle-type identified using clustering algorithm applied to all combined samples.**

**A picture containing chart

Description automatically generated**As a complementary analysis method, the rule-based classification scheme separates the particles into five different operator defined categories: “Sea Salt,” “Sea Salt/Sulfate,” “Sulfates/Organics,” “Carbonaceous,” and “Other.” Particle categories are first classified according to the Na content where if Na is greater than or equal the threshold value of 1%. These particles are further divided into separate classes of [Na] > [S] classified as “Sea Salt”, whereas those of [Na] < [S] classified as “Sea Salt/Sulfate.” Particles of [Na] < 1% are further classified as “Carbonaceous” if concentration of other elements excluding Na, Cl, S is < 1%; Particles with [C, N, O, S] ≥ 0.5% are classified as “Carbonaceous/Sulfate.” Particles not classified into either of the above four classes are placed into a single nonspecific category “Other”. The reported particle size used in this work is based on projected area equivalent diameter (AED, µm), which assumes a 2D projected image of the equivalent circle diameter of a particle silhouette as it is readily available from SEM data.

**Figure S6. A) Classification scheme applied to particles from aircraft collected samples; B) Mean elemental contribution for each particle-type using rule-based method.**

**A picture containing graphical user interface

Description automatically generatedFigure S7. CCSEM size distribution rule-based particle classification scheme with average FIMS size distribution across different synoptic condition (non-dry intrusion vs. dry intrusion) and atmospheric layer (free troposphere vs. marine boundary layer) fitted with lognormal mode diameter (grey dashed lines). The CCSEM size distribution is superimposed and anchored at 0.25 µm as a visual comparison. The composition of the size-segregated particle-type population were broken down into “Carbonaceous” (i.e., Carbonaceous + Carbonaceous/Sulfate), “Inorganics” (i.e., Sea Salt + Sea Salt/Sulfate), and “Other”.**

**Chart, bar chart

Description automatically generatedFigure S8. Particle-type fraction detected at different atmospheric layer (MBL vs. FT) and synoptic condition (non-DI vs. DI) for different research flights based on k-means clustering.**

**A screenshot of a computer

Description automatically generatedFigure S9. Collage of singular value decomposition map based on the STXM/NEXAFS imaging of particles in all analyzed samples collected at different atmospheric layer and synoptic conditions. Areas dominated by elemental carbon/soot components are shown in red, inorganic dominated regions are blue, and regions containing organic constituents are green. Note that components can overlap where each pixel can contain different combination of the individual components: EC + IN constituents as purple; OC + EC as yellow; OC + IN as cyan.**

**Diagram, schematic

Description automatically generatedFigure S10. Size-resolved organic volume fraction ratio measured by STXM for individual particles superimposed with the particle size distribution measured by FIMS onboard the G-1 aircraft.**

|  |  |  |  |
| --- | --- | --- | --- |
|  | | | |
| **Component type** | **OVF Integrated Area**  **FT non-DI** | **OVF Integrated Area**  **FT DI** | **% Increase** |
| NaCl-Sucrose | 225.61 | 314.55 | 39.42 |
| NaCl-Adipic acid | 228.40 | 316.92 | 38.76 |
| NaCl-Glucose | 234.18 | 337.20 | 43.99 |
| NaCl-Oxalic acid | 262.04 | 379.73 | 44.91 |
| (NH4)2SO4-Sucrose | 151.64 | 158.16 | 4.30 |
| (NH4)2SO4-Adipic acid | 152.39 | 159.06 | 4.38 |
| (NH4)2SO4-Glucose | 155.99 | 163.37 | 4.73 |
| (NH4)2SO4-Oxalic acid | 168.80 | 188.17 | 11.48 |
| **Component type** | **OVF Integrated Area**  **MBL non-DI** | **OVF Integrated Area**  **MBL DI** | **% Increase** |
| NaCl-Sucrose | 356.48 | 921.19 | 158.41 |
| NaCl-Adipic acid | 358.14 | 915.13 | 155.52 |
| NaCl-Glucose | 364.93 | 959.31 | 162.88 |
| NaCl-Oxalic acid | 390.86 | 1138.52 | 191.28 |
| (NH4)2SO4-Sucrose | 246.79 | 426.69 | 72.89 |
| (NH4)2SO4-Adipic acid | 247.59 | 430.65 | 73.94 |
| (NH4)2SO4-Glucose | 254.13 | 465.22 | 83.07 |
| (NH4)2SO4-Oxalic acid | 276.98 | 571.60 | 106.37 |
|  | | | |

**Chart

Description automatically generatedTable 2. Calculated integrated area of STXM-derived organic volume fraction (OVF) for different inorganic-organic components. Relevant inorganic-organic components were chosen based on relevant particle-types in the region (i.e. marine environment) and previous literature used to calculate OVF (Fraund et al., 2019; Pham et al., 2017): Inorganic – NaCl, (NH4)2SO4, Organic – sucrose, adipic acid, glucose, and oxalic acid. The integrated area was calculated by taking the sum of the percent contribution as a function of OVF as shown in Figure 6 applied for the different combinations of inorganic-organic components.**

**Figure S11. CCN activity of individual particles. A) Average FIMS particle size distribution for selected episodes during the ACE-ENA experiment. B) Comparison of measured and calculated CCN concentrations at different supersaturation (SS) values (0.13%, 0.14%, and 0.15% SS).**

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