

Halogen Production from Playa Dust Emitted from the Great Salt Lake: Implications of the Shrinking Great Salt Lake on Regional Air Quality

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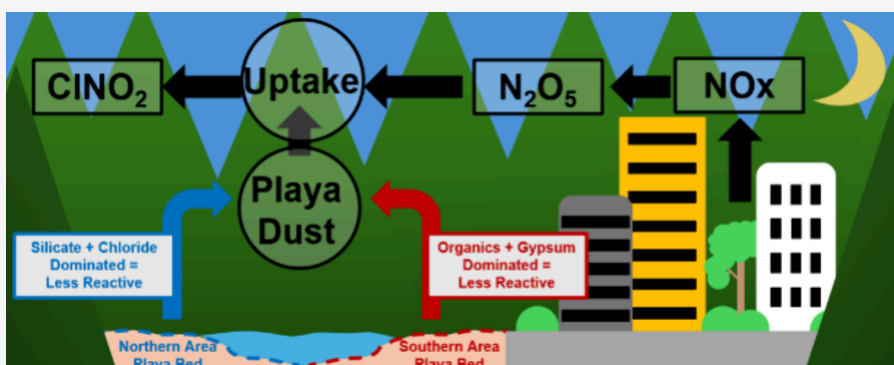
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ABSTRACT: Halogen radicals, such as atomic chlorine ($\text{Cl}\cdot$), can contribute to secondary wintertime fine particulate matter in the Salt Lake Valley. One source of $\text{Cl}\cdot$ is the photolysis of nitryl chloride (CINO_2), formed from the reaction of dinitrogen pentoxide (N_2O_5) with chloride-containing aerosol. However, sources of chloride-containing aerosols in the Salt Lake Valley, and their subsequent reaction kinetics, remain poorly constrained. We analyzed playa (i.e., dried saline lakebed) samples collected from dust-emitting regions along the northern and southern areas of the shrinking Great Salt Lake to investigate their mineralogy, reactivity, and CINO_2 forming potential. The reactive uptake coefficients ($\gamma_{\text{N}_2\text{O}_5}$) for all samples ranged from 0.005 to 0.064, with the average $\gamma_{\text{N}_2\text{O}_5}$ of the northern area samples approximately double the average $\gamma_{\text{N}_2\text{O}_5}$ of the southern area samples. We attribute the increased $\gamma_{\text{N}_2\text{O}_5}$ of northern playas to increased particulate chloride and silicate, while the reduced $\gamma_{\text{N}_2\text{O}_5}$ in southern playas is due to particulate organics and high quantities of gypsum, a nonreactive mineral. The yield of CINO_2 is $> 50\%$ for all playas tested, with one exception. Using our kinetic data during an ambient wintertime case study, we estimate playa dust contributes up to 5% of observed CINO_2 , a lower estimate which likely increases during the spring when dust emissions are higher. Our work highlights the importance of including playa dust in current air quality models, especially as reductions of anthropogenic halogen sources are implemented in the United States, and ephemeral lakes continue to shrink globally.

KEYWORDS: aerosol, air quality, halogen production, mineral dust, nitryl chloride, dinitrogen pentoxide

1. INTRODUCTION

In the past 40 years, the Great Salt Lake has shrunk by nearly 50%, exposing approximately 2000 km^2 of lakebed.^{1–4} One consequence of the lake shrinking is an increased exposure of playa (dried saline lakebed) for dust production^{5–8} and transport across the Salt Lake Valley.⁹ This region also experiences severe particulate matter (PM) pollution events due to accumulation of fine ammonium nitrate (NH_4NO_3) on aerosol, which become trapped near the valley floor by persistent cold air pools (PCAPs).^{10–17} The buildup of wintertime NH_4NO_3 in the valley has been the subject of several recent field campaigns, including the 2017 Utah Winter

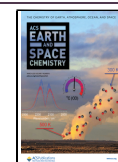
Fine Particulate Study (UWFPS), and its formation has been shown to be nitric acid (HNO_3)-limited.^{18,19} A key question of UWFPS is what sources of oxidants contribute to nitric acid formation and play a role in the formation of secondary wintertime air pollutants.²⁰ Current air quality models consider

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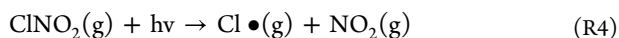
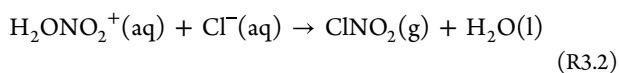
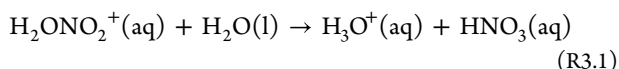
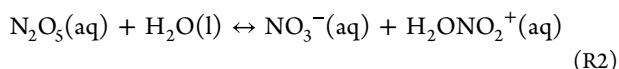
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playa dust as a source of coarse PM, but do not consider playa dusts' ability to participate in chemical reactions that can degrade air quality and potentially contribute to the oxidant budget beyond serving as a sink of acidic gases, including HNO_3 , which would suppress the formation of NH_4NO_3 .^{21–26} Previous studies have demonstrated that playas can potentially contribute to the oxidant budget by heterogeneously reacting with dinitrogen pentoxide (N_2O_5), a common reservoir for nitrogen oxide radicals ($\text{NO}_x \equiv \text{NO} + \text{NO}_2$), by generating nitryl chloride (ClNO_2) due to the high halide content of playa dusts.^{27,28} ClNO_2 photolyzes to produce the chlorine radical ($\text{Cl}\bullet$), a powerful atmospheric oxidant that has been shown to react with volatile organic compounds (VOCs) and facilitate the formation of ozone (O_3)²⁹ and the peroxy radical ($\text{RO}_2\bullet$),³⁰ both of which contribute to the formation of HNO_3 via the oxidation of NO_x , and later NH_4NO_3 in the Salt Lake Valley.^{10,21,25} However, we lack knowledge about the reactivity and halogen generating potential of dust-emitting regions around the Great Salt Lake, limiting our ability to add this potentially important oxidant source to regional air quality models.

A reaction pathway leading to the formation of $\text{Cl}\bullet$ starts when N_2O_5 reacts with a particle (R1–R2). The efficiency of this heterogeneous reaction is referred to as the reactive uptake coefficient ($\gamma_{\text{N}_2\text{O}_5}$).³¹ N_2O_5 will either undergo a hydrolysis reaction to produce aqueous NO_3^- R3.1,^{32–34} or, in the presence of aqueous particulate chloride ($\text{Cl}^-_{(\text{aq})}$), N_2O_5 will react to form gaseous ClNO_2 R3.2.^{34–38} The amount of ClNO_2 produced per molecule of N_2O_5 reacted is denoted as the yield of ClNO_2 (ϕ_{ClNO_2}). In the daytime, ClNO_2 will photolyze to regenerate $\text{NO}_x(\text{g})$ and form $\text{Cl}\bullet$ R4, increasing the oxidant budget.^{31,39}



Previous work has also shown that the rate constant of R3.2 is 400–1000 times faster than R3.1, which suggests that ClNO_2 can still form even in the presence of low concentrations of $\text{Cl}^-_{(\text{aq})}$.^{31,35,37,40,41}

ClNO_2 is known to be prevalent in inland regions but the sources of aerosol chloride leading to its formation are poorly characterized.^{21,38,42,43} Potential inland aerosol sources include biomass burning,^{44,45} road salt,^{46,47} coal fired power plants,^{48,49} industrial activity,⁵⁰ and playa dust.^{27,28} However, of these inland sources, only biomass burning and industrial sources are included in current air quality and chemical transport models that parameterize reactive chlorine chemistry.^{51–53} In addition to an incomplete inventory of aerosol chloride sources in inland regions, barriers to incorporating other sources in air quality models are a lack of known reactive uptake kinetics and product yields (i.e., $\gamma_{\text{N}_2\text{O}_5}$ and ϕ_{ClNO_2}) for relevant sources.⁴¹

Given the reduction in anthropogenic halogen emissions as the United States transitions to renewable energy sources,^{54–56}

understanding the importance of chloride-containing aerosols from natural sources, such as playa dust, is critical, particularly with playa dust sources increasing as saline lakes continue to shrink both at the Great Salt Lake and worldwide.^{3,56,57} Playa dust emissions are especially important in the Salt Lake Valley as it is proximal to multiple playa sources, including the Great Salt Lake, producing large quantities of saline-rich dust in the spring and summer months.^{2,9,58,59}

To better understand potential, current, and future impacts of the shrinking Great Salt Lake on reactive halogen chemistry, we quantified $\gamma_{\text{N}_2\text{O}_5}$ and ϕ_{ClNO_2} using aerosol generated from playa samples collected from major dust emitting regions around the Great Salt Lake. We quantified the N_2O_5 uptake and ClNO_2 formation under varying relative humidities to simulate different atmospheric conditions. Further, we performed single particle and bulk chemical analyses of the generated aerosol to link particle composition and dust mineralogy with their ClNO_2 formation potential. As such, we provide the chemical and kinetic information needed to include playa dust aerosol in parameterizations of the N_2O_5 – ClNO_2 chemical mechanism in the Salt Lake Valley. We extend our laboratory results to estimate the contribution of playa dust to ClNO_2 production during the 2017 UWFPs campaign.

2. METHODS

2.1. Generation of Playa Dust Aerosol. Playa samples were obtained from eight dust emitting regions around the Great Salt Lake with samples evenly spread across the Northern Area (NA) and Southern Areas (SA) of the lakebed, which are separated by a causeway at the center of the lake. A labeled map is available in Figure S1 of the Supporting Information (SI). Coordinates and the collection date of these samples are also available in Table S1. All playa sediment samples were used “as received”, with a brief description of the collection and processing method available in Section 1 of the SI.⁷ The playa sediment samples were aerosolized in the laboratory and therefore represent “freshly emitted” dust. A custom dry aerosol generation setup was used to generate playa dust aerosol.²⁷ This method utilizes a wrist-action shaker to agitate sediment samples in a custom-designed Erlenmeyer flask, which simulates natural dust emissions through saltation, sandblasting, and entrainment. Once airborne, the dust is transported to a dilution drum using ultrahigh purity N_2 as a carrier gas, which reduces spikes in aerosol concentration. The resulting flow passes through a cyclone (Mesalabs; Model SCC 2.229) with a PM_{10} cutoff to remove supermicron particles. The PM_{10} cutoff was chosen to provide a stable surface area concentration (Sa); however, we do note that the PM_{10} cutoff likely has a minor effect on soluble ion content.⁶⁰ The PM_{10} cutoff can also be used to represent particles that are susceptible to long-range transport, which likely have a larger spatial impact on halogen activation.⁶¹ However, previous observations of ambient mineral dust suggests that the particle size ranges from 0.1 to $10\ \mu\text{m}$,^{2,62} so we are likely only partially covering ambient dust sizes observed in the Salt Lake Valley. The size-selected dust was neutralized by two ^{210}Po strips (NRD, LLC; Model 2U500) to reduce aerosol losses within the system.^{27,63} Afterward, the dust sample was introduced to a humidified dilution flow of ultrahigh purity N_2 ($\sim 5\ \text{L min}^{-1}$) to condition the relative humidity (RH) of the aerosolized particles. The RH was measured using a relative humidity probe (model HMP60, Vaisala) before entering an aerosol

flow tube (AFT). To ensure RH remains consistent throughout the AFT, the RH was periodically measured at the exit to the flow tube. The humidity of the AFT for our experiments ranged from 30 to 45% RH, where 45% RH was the upper limit of what was achievable with these samples.

2.2. Aerosol Flow Tube, Kinetic Measurements of N_2O_5 , ClNO_2 , and Particle Surface Area. The generated playa dust aerosol orthogonally entered a 90 cm long, 6 cm inner diameter, Pyrex AFT coated with halocarbon wax (Halocarbon Corp.), described in Figure S2. N_2O_5 was generated continuously in a Teflon vessel through the formation of the nitrate radical (NO_3) via the reaction of nitrogen dioxide (NO_2) (10 ppm standard; Airgas) and O_3 , which was generated with a UV-pen lamp (Jetlight Model 610) and ultrapure zero air. NO_3 was further reacted with excess NO_2 to produce N_2O_5 in equilibrium at room temperature.^{31,64,65} The final flow was $\sim 0.1 \text{ L min}^{-1}$ and N_2O_5 was introduced to the AFT through a Teflon lined stainless steel movable injector along the center of the AFT. A collision limit calculation,⁶⁶ which is described in Section 2.1 of the SI, was used to estimate that ~ 2.0 ppb of N_2O_5 was introduced to the AFT during our experiments.

The output of the flow tube entered the iodide-adduct chemical ionization mass spectrometer (CIMS)⁶⁴ through a critical orifice at $\sim 2 \text{ L min}^{-1}$ of flow. N_2O_5 and ClNO_2 were detected as $[\text{I} \cdot (\text{N}_2\text{O}_5)]^-$ (234.9 amu) and $[\text{I} \cdot (\text{ClNO}_2)]^-$ (207.9 and 209.9 amu) with isotope ratios confirming our ion peak assignments.⁶⁴ The humidified total particle surface area was measured simultaneously at the exit of the AFT using both a scanning mobility particle sizer (SMPS; model 3082, TSI, Inc.) and an aerosol particle sizer (APS; model 3321, TSI, Inc.), which ranged from 3.2×10^{-5} to $2.8 \times 10^{-4} \text{ cm}^2/\text{cm}^3$ for all experiments. Example particle surface area distributions are also available in Figure S3 of the Supporting Information. To keep laminar flow in the AFT, the APS sample flow was diluted by a factor of ~ 10 using ultrahigh purity N_2 . We also sampled the flow exiting the AFT with a single particle mass spectrometer (see Section 2.4) to determine the size and chemical composition of the generated aerosols. The total flow of the Single Particle Mass Spectrometer, CIMS, SMPS, and APS together was $\sim 3 \text{ L min}^{-1}$, with a mean residence time of $\sim 15 \text{ s}$ in the flow tube.

The CIMS utilizes methyl iodide (CAS #74–88–4) to generate iodide-adducts, which is considered toxic and carcinogenic. To reduce any hazards associated with the use of methyl iodide, the authors stored the methyl iodide in an airtight chamber to generate I^- and handled the chemical under a fume hood while wearing laboratory coats, chemical resistant butyl gloves, and laboratory goggles.

2.3. Determination of $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 . To determine $\gamma\text{N}_2\text{O}_5$, we used particle modulation, an appropriate method for dry aerosol generation,^{27,31} where the injector is at a fixed position (40 cm from the top of the AFT). The dust output enters the flow tube at different surface area concentrations in $\sim 15 \text{ min}$ intervals. This allowed us to develop $\sim 10 \text{ min}$ of steady state conditions in the reaction zone, where the aerosol and N_2O_5 interact. The N_2O_5 signal was determined both with and without aerosols to determine the pseudo-first-order rate constant for N_2O_5 loss (k_{het}), which is calculated using eq 1:^{27,31}

$$k_{\text{het}} = -\left(\frac{1}{\Delta t}\right) \ln \left(\frac{[\text{N}_2\text{O}_5]_{\text{w/particles}}}{[\text{N}_2\text{O}_5]_{\text{wo/particles}}} \right) \quad (1)$$

where $[\text{N}_2\text{O}_5]_{\text{w/particles}}$ is the signal when particles are present and $[\text{N}_2\text{O}_5]_{\text{wo/particles}}$ is the signal when particles are not present. k_{wall} , calculated in eq 2, measured the change in N_2O_5 at the top and bottom positions of the flow tube in the absence of dust particles:^{27,40,67,68}

$$k_{\text{wall}} = -\left(\frac{1}{\Delta t}\right) \ln \left(\frac{[\text{N}_2\text{O}_5]_{\text{top}}}{[\text{N}_2\text{O}_5]_{\text{bottom}}} \right) \quad (2)$$

where Δt is the reaction time (dependent on the injector position within the flow tube) in s, $[\text{N}_2\text{O}_5]_{\text{bottom}}$ is the signal when the injector is at the bottom position, and $[\text{N}_2\text{O}_5]_{\text{top}}$ is the signal when the injector is at the top position. Measurements of $[\text{N}_2\text{O}_5]_{\text{top}}$ and $[\text{N}_2\text{O}_5]_{\text{bottom}}$ were taken in $\sim 10 \text{ min}$ intervals. This allowed $\sim 5 \text{ min}$ to establish laminar flow after the injector was moved between 40 cm (top position) and 20 cm (bottom position). $\gamma\text{N}_2\text{O}_5$ was then calculated using eq 3:

$$\gamma\text{N}_2\text{O}_5 = \frac{4(k_{\text{het}})}{\omega S_a} \quad (3)$$

where ω is the mean molecular speed of N_2O_5 in cm/s (held constant at $2.4 \times 10^4 \text{ cm/s}$ assuming a temperature of 295 K) and S_a is the total surface area concentration in cm^2/cm^3 . Each experiment was repeated 5 times at every tested humidity which produced linear correlations between k_{het} and S_a ($R^2 = 0.92$, Figure S4). We also validated our reactive kinetics measurements using full particle decays and pseudoparticle modulation on some of our playa dust samples. All measured values of S_a were corrected for aerosol density and shape (see Table S2),⁶⁹ which is detailed in Section 3.1 of the SI. The $\gamma\text{N}_2\text{O}_5$ values were also corrected for nonplug flow conditions from the observed k_{wall} ^{31,70} and gas-phase diffusion limitations.⁷¹ The experimental setup was periodically calibrated using commercially available sodium chloride (NaCl, Sigma-Aldrich $\geq 99\%$ Purity), which was suspended via an atomizer to generate aerosols of a known reactivity with N_2O_5 (~ 0.03) and produced ClNO_2 with a yield of ~ 1 (SI Section 2.1).³⁶

ϕClNO_2 was validated by performing routine calibration for the sensitivity of ClNO_2 relative to N_2O_5 by flowing N_2O_5 over a wetted NaCl slurry, which theoretically produces a 1:1 yield of ClNO_2 .⁶⁴ Sensitivity tests were done at RH values of 30, 40, 45, and 50% to match our experimental conditions, and were then used to calculate the observed ϕClNO_2 described in eq 4:

$$\phi\text{ClNO}_2 = \frac{[\text{ClNO}_2]_{\text{w/particles}} - [\text{ClNO}_2]_{\text{wo/particles}}}{[\text{N}_2\text{O}_5]_{\text{wo/particles}} - [\text{N}_2\text{O}_5]_{\text{w/particles}}} \quad (4)$$

where $[\text{ClNO}_2]_{\text{w/particles}}$ is the ClNO_2 signal in the flow tube when particles are present and $[\text{ClNO}_2]_{\text{wo/particles}}$ is the background ClNO_2 signal in the flow tube.

2.4. Single Particle Analysis. At the exit of the AFT, the size-resolved chemical composition of both reacted and unreacted dust particles were also determined using a Single Particle Aerosol Mass Spectrometer (SPMS; Model 0515, Hexin Inc.).⁷² The SPMS operates by sampling aerosols through an aerodynamic lens resulting in a single particle beam. Particles are accelerated to their size-dependent terminal velocity, which are scattered by two 532 nm continuous wave

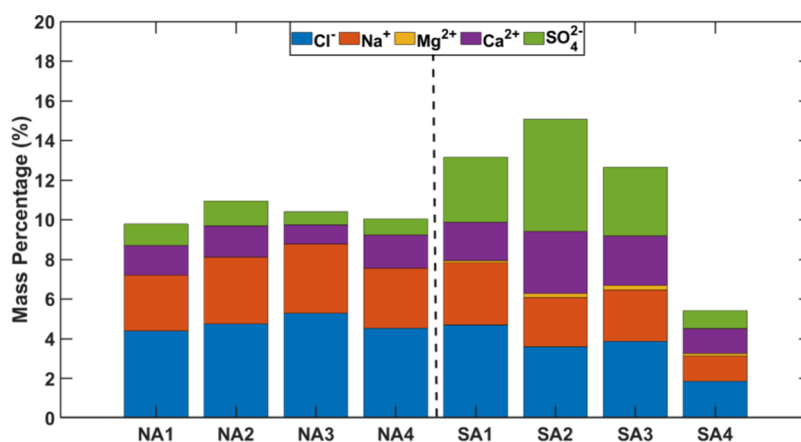


Figure 1. Mass percentage of soluble ions measured for each of the collected playa samples. The dashed line delineates samples from the Northern Area (NA) of the lake and the Southern Area (SA) of the lake.

lasers (CNI, MLL-532–50mW) that are 6 cm apart. The timing of these two scatters by each particle, as measured by a PMT matched to each laser, enables calculation of the individual terminal velocity, which is then converted to the vacuum aerodynamic diameter (d_{va}) by calibration with polystyrene latex spheres (PSLs) of a known size. The particle sizing region also initiates a timing circuit, which times the firing of a 266 nm 20-Hz Nd:YAG laser (Quantel Ultra 50). For this study, the 266 nm laser was operated at ~ 0.5 mJ/pulse to induce laser desorption/ionization producing both positive and negative ions from the individual particles, which were detected using a dual polarity time-of-flight mass spectrometer.

Approximately 1500 particles from each experiment were analyzed with SPMS, and the dual-polarity single-particle mass spectra were imported into MATLAB (ver. 2021b; The Mathworks Inc.) and analyzed using the Flexible Analysis Toolkit for the Exploration of Single-particle mass spectrometer data (FATES).⁷³ The individual mass spectra were clustered based on the presence and intensity of ion peaks using an Adaptive Resonance Theory-based neural network algorithm (ART-2a) at a vigilance factor of 0.8.⁷⁴ These clusters were then merged and compared to literature of single particle data on dust and other single particle signatures.^{28,75,76}

2.5. Predicting Playa Mineralogy using ISORROPIA II.

Thermodynamic modeling was used to predict the mineral composition of the playa dust evaporites (e.g., saline material) using soluble ion concentrations measured by ion chromatography (IC, method described in Section 3.2 of the SI). We utilized ISORROPIA-II,⁷⁷ which was run at an RH of 5% to predict the mineralogy in stable mode. By running the model in stable mode, we allow salts to precipitate under supersaturated conditions. The model estimates concentrations of halite (NaCl), potassium chloride (KCl), magnesium chloride (MgCl), and calcium sulfate (CaSO_4), which we assume are in their more stable, hydrated, forms. Excess, unbalanced Ca^{2+} is assumed to be in the form of calcite (CaCO_3).^{75,78}

2.6. Estimating ClNO_2 Production from Playa Dust during the UWFPS Campaign. We extended our laboratory kinetics measurements to estimate the contribution of ClNO_2 from Great Salt Lake playa dust during the 2017 Utah Wintertime Fine Particulate Study (UWFPS), which included measurements of particle surface area concentration, soluble ions indicative of playa evaporites, N_2O_5 , and ClNO_2 .²⁰ The production of ClNO_2 (P_{ClNO_2}) in pptv/s was estimated using eq 5:⁷⁹

$$P_{\text{ClNO}_2} = \phi \text{ClNO}_2(t) \gamma \text{N}_2\text{O}_5(t) [\text{N}_2\text{O}_5](t) \left\{ \frac{\text{Sa}(t) \omega(t)}{4} \right\} \quad (5)$$

where $[\text{N}_2\text{O}_5](t)$ represents the estimated concentration of N_2O_5 in pptv at a given time, $\text{Sa}(t)$ is the total surface area concentration of playa dust in cm^2/cm^3 at a given time, $\omega(t)$ represents the mean molecular speed adjusted for ambient temperature during the field campaign in cm/s, and both $\gamma \text{N}_2\text{O}_5$ and ϕClNO_2 represent the reactive uptake of N_2O_5 and subsequent yield of ClNO_2 derived from our laboratory measurements of playa dust.

To estimate P_{ClNO_2} , we utilized coarse mode ($\text{PM}_{10} - \text{PM}_{2.5}$) $\text{Sa}(t)$ measurements collected every 5 min from an APS at the University of Utah Field Site. During UWFPS, dust contributed to an estimated 5% of the total coarse mode surface area concentration measured during PCAP events in the Salt Lake Valley, which occurred from 01/30/2017–02/02/2017.¹⁸ To ensure that the likely source of the mineral dust is from the Great Salt Lake, we performed a 6 h back trajectory model every hour throughout the PCAP periods observed during the campaign using the NOAA Hybrid Single-Particle Lagrangian Integrate Trajectory (HYSPLIT) model shown in Figure S5.⁸⁰ We chose to use the North American Mesoscale 12 KM Forecast System (NAM 12KM) for our meteorology data set as it has a high enough resolution to predict regional air mass back trajectories in the Salt Lake Valley. The back trajectory overlaps with the Great Salt Lake during the first PCAP, suggesting the dust sourced during this period is likely from the Great Salt Lake. We do note that the model was initialized at 1000 m A.G.L to avoid any effects that topography may have on the model. We also assume that the dust sourced from the Great Salt Lake is evenly distributed between the Northern and Southern Areas of the lake.

The UWFPS campaign also had measurements of ambient ClNO_2 and N_2O_5 concentrations at the Logan Field Site using an iodide-adduct high-resolution time-of-flight Chemical Ionization Mass Spectrometer (I^- HR-ToF CIMS).²⁰ This field site is located approximately 100 km north of the University of Utah field site, and, relative to Salt Lake City, experiences more severe $\text{PM}_{2.5}$ and PM_{10} pollution under inversion conditions.⁸¹ Observations of wintertime N_2O_5 in the Salt Lake Valley vary greatly and are largely dependent on inversion versus noninversion conditions. The Logan Field Site observation of nighttime N_2O_5 under PCAP conditions were low, but detectable (<50 pptv).²⁰ The low observed

concentrations of N_2O_5 at the Logan Field Site during UWFPs can likely be attributed to elevated ambient aerosol concentrations reacting with N_2O_5 and increased concentrations of nitrogen oxide (NO) that can titrate O_3 and N_2O_5 during PCAP events. Previous observations of N_2O_5 in Salt Lake City during the winter estimate ambient N_2O_5 concentrations range from 300–1500 pptv during non-inversion wintertime conditions in the valley, which decrease to ~ 100 pptv under PCAP conditions.¹⁰ Given the wide range of observed concentrations of N_2O_5 around the Great Salt Lake, we estimate P_{CINO_2} when $[\text{N}_2\text{O}_5](t)$ is 50 pptv, 100 pptv, 800 pptv, and 1000 pptv. Calculations of P_{CINO_2} when $[\text{N}_2\text{O}_5](t)$ is 50 and 100 pptv represent the expected ambient concentration of N_2O_5 under PCAP conditions. Calculations of P_{CINO_2} when $[\text{N}_2\text{O}_5](t)$ is 800 and 1000 pptv represent noninversion conditions, when particulate matter and NO concentrations are expected to be lower.

3. RESULTS AND DISCUSSION

3.1. Differences in Playa Evaporite Composition between the Northern and Southern Areas of the Great Salt Lake. Figure 1 depicts the percent contribution by weight of soluble inorganic ions from IC analysis for each of the playa samples. Table S3 also includes soluble ion concentrations of lake water from the Northern and Southern Areas of the Great Salt Lake (collection method for water samples are described in Section S1 of the SI). The most striking difference between playa samples from the Northern and Southern Areas of the lake is the variation in soluble calcium (Ca^{2+}) and sulfate (SO_4^{2-}). Playas from the Northern Area of the lake contain soluble Ca^{2+} and SO_4^{2-} ranging from 9.71 ± 0.09 to 16.8 ± 0.2 g/kg and 6.6 ± 0.4 to 12.6 ± 0.3 g/kg, respectively. In contrast, playas in the Southern Area of the lake show 1.5- to 3.5-fold higher observed Ca^{2+} and SO_4^{2-} concentrations, ranging from 12.5 ± 0.1 to 31.2 ± 0.6 g/kg and 9.1 ± 0.2 to 56.6 ± 0.4 g/kg, respectively. Our results suggest an increased presence of calcium and sulfate containing minerals, such as gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), are present in playas from the Southern Area of the lake.

Concentrations of sodium (Na^+) are significantly higher ($p \leq 0.05$) in playas collected from the Northern Area of the lake (28.2 ± 0.1 to 35.0 ± 0.3 g/kg) when compared to playa collected from the Southern Area (12.6 ± 0.1 to 31.4 ± 0.2 g/kg). A similar trend is observed with chloride (Cl^-) concentrations, which are approximately 1.5 times higher in the Northern Area playa samples (43.9 ± 0.3 to 52.8 ± 0.2 g/kg) when compared to the Southern Area (18.5 ± 0.2 to 46.9 ± 0.2 g/kg). This increase in observed Na^+ and Cl^- in playa samples from the Northern Area of the lake can be attributed to increased concentrations of Na^+ and Cl^- in water samples (1217 ± 2 and 2493.0 ± 0.8 g/L, respectively), which is approximately 3 times higher than the observed concentrations of Na^+ and Cl^- in water collected from the Southern Area of the lake (378 ± 2 and 715.2 ± 0.5 g/L, respectively). The difference in salinity is consistent with previously observed differences in water salinity between the Northern and Southern areas of the lake.⁸² The increase in water salinity likely leads to higher concentrations of evaporites present in the playa samples,^{83–87} such as halite (NaCl), in our Northern Area playa samples. The higher salinity in the Northern Area of the lake has widely been attributed to the installation of the causeway in the late 1950s (separating the Northern and Southern areas of the lake), which reduces the mixing of water

between the Northern and Southern areas of the lake,^{82,88} and restricts freshwater inflow from tributary rivers to only the Southern Area of the Great Salt Lake.⁸⁹

Despite measuring appreciable concentrations of magnesium (Mg^{2+}) in water collected from the Northern Area of the lake (87 ± 15 g/L), we measured unexpectedly low concentrations of Mg^{2+} in playa samples collected from the Northern Area of the lake, with all playa samples measuring below the detection limit for Mg^{2+} . However, trace levels of magnesium are present, as our single particle data show characteristic peaks for magnesium ($^{24}\text{Mg}^+$, Figure S6). In contrast, the concentration of Mg^{2+} in the water collected from the Southern Area is below the detection limit, which is likely attributed to a reduction in salinity from the nearby tributary rivers.^{82,88,90} The playas collected from the Southern Area of the lake do have observable concentrations of Mg^{2+} (1.10 ± 0.02 – 2.38 ± 0.04 g/kg), which is likely due to the presence of soluble magnesium-containing minerals.

To better understand our soluble ion data, we used ISORROPIA-II to predict the types of evaporites present in our samples. As shown in Table S4, the primary evaporites present in all samples are NaCl and calcium sulfate (CaSO_4), which is likely present in the more stable, hydrated form of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) based on previous observations of playa beds.^{91–95} The most prominent mineral predicted in the Northern Area of the lake is NaCl , which has an evaporite percent composition greater than 80% by mass for all Northern samples. In contrast, the predicted mineralogy for the Southern Area of the lake is dominated by a mix of CaSO_4 and NaCl , where the NaCl percent composition is predicted to be less than 65% by mass and CaSO_4 ranges from 26% to 57% by mass, which is much higher than the predicted abundance of CaSO_4 in the Northern Area of the lake (9–18% by mass). The higher abundance of predicted NaCl in the Northern Area of the lake is likely due to the increased salinity of lake water when compared to the Southern Area.^{88,90,96} Several samples also contained unbalanced Ca^{2+} . Since carbonate-containing compounds have previously been suggested to be an important component of playa dusts,^{75,97,98} coupled with the presence of characteristic peaks observed on the SPMS ($^{56}\text{CaO}^+$ and $^{96}\text{Ca}_2\text{O}^+$, Figure S6),^{75,99} we suggest that the unbalanced Ca^{2+} is present in the form of CaCO_3 (Table S4). The estimated concentration of CaCO_3 ranges from 2.00 ± 0.02 – 3.30 ± 0.01 $\mu\text{mol}/\text{m}^3$, with the largest concentration of calcite present in sample NA-4.

3.2. How Differing Mineralogy between the Northern and Southern Areas of the Great Salt Lake Affects CINO_2 Yields. Samples from the Northern and Southern Areas of the lake show similar trends in yields of CINO_2 despite their different mineralogy (see Figure 2 and Table 1). For all samples, with the exception of Northern Area-Sample 2 (NA-2) ($\varphi\text{CINO}_2 = 0.38 \pm 0.06$), playa dusts from the Northern Area and Southern Area produced high yields of CINO_2 at an RH of 30% (φCINO_2 varied between 0.79 ± 0.14 and 1.16 ± 0.16). We find the high yields at relatively low RH values ($\sim 30\%$) surprising as the mineralogy predictions from ISORROPIA-II (Table S4) and ion concentrations from IC (Figure 1) suggest that halite is the primary evaporite source in our playa dust, followed by gypsum. If halite were the only chloride-containing evaporite present in our samples, CINO_2 would likely not be produced at any tested RH since pure NaCl deliquesces at 75% RH.^{100–102} This is because our samples are generated dry, then humidified, rather than

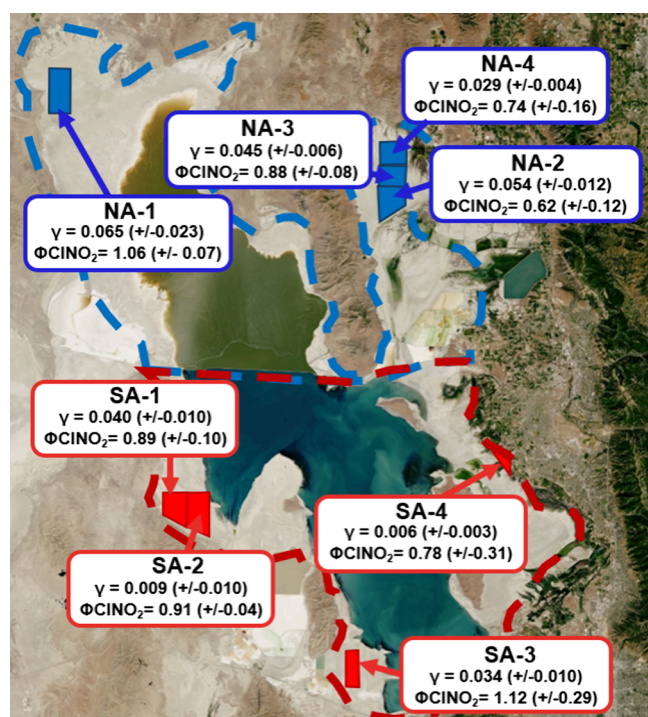


Figure 2. Measured $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 values for aerosol generated from the collected Great Salt Lake playa samples at an RH of 40%. The red and blue outlines show exposed playa from reduction in water level over the past 40 years,⁵⁶ highlighting the increase in new playa sources. The area outlined in blue represents the Northern Area of the Great Salt Lake (Gunnison Bay/Bear River Bay), while the area outlined in red is representative of the Southern Area of the lake (Gilbert Bay/Farmington Bay). Error is characterized as 95% confidence intervals. The background map image is provided by the NASA Earth Observatory and U.S. Geological Survey Landsat data set.

undergoing wet generation followed by drying to reach the desired humidity.⁶⁰ The high yields also do not vary significantly when RH increases, unlike some of the playa samples measured from different regions of the south and southwestern U.S.²⁷ Our results suggest the presence of chloride-containing evaporites other than halite, similar to previous playa studies.²⁸ Notably, the single particle mass spectrometry measurements supports the presence of chloride-containing ions other than halite—including $^{129,131,133}\text{MgCl}_3^-$, $^{127,129,131}\text{CaCl}_2\text{OH}^-$, and $^{59}\text{MgCl}^+$ —that likely are hydrated and play an important role in providing enough water to initiate the uptake of N_2O_5 and formation of ClNO_2 . The identity of these evaporites are likely hygroscopic salts with a low deliquescence relative humidity (DRH), such as $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ (DRH = 33%)¹⁰³ or $\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$ (DRH = 12–20%),¹⁰⁴ which have previously been suggested to facilitate ClNO_2 formation at trace concentrations.²⁸

We further probe this hypothesis by comparing the reactive uptake coefficient of N_2O_5 and yield of ClNO_2 using MgCl_2 standards, in addition to the more studied NaCl standard.³¹ As shown in Figure S7 and Table S5, MgCl_2 can react efficiently with N_2O_5 even at a relatively low RH (RH = 20%) while producing a 1:1 yield of ClNO_2 , unlike NaCl , which effloresces at a high humidity (ERH = 45%)¹⁰⁵ and is, therefore, unreactive at lower humidities.¹⁰⁶ The production of ClNO_2 from MgCl_2 at a low RH is not surprising given that MgCl_2 can retain water at humidities as low as 3%,¹⁰⁷ emphasizing the

Table 1. Laboratory Measurements of $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 at Varying Relative Humidities (RH)^a

Sample	RH	$\gamma\text{N}_2\text{O}_5$	ϕClNO_2
NA-1	30	0.043(±0.011)	1.01 (±0.11)
	40	0.065(±0.023)	1.06 (±0.07)
	45	0.036(±0.004)	1.12 (±0.01)
NA-2	30	0.042 (±0.006)	0.38 (±0.05)
	40	0.054 (±0.012)	0.62 (±0.12)
	45	0.064 (±0.011)	0.72 (±0.08)
NA-3	30	0.029 (±0.005)	0.95 (±0.04)
	40	0.045 (±0.006)	0.88 (±0.08)
	45	0.045 (±0.006)	1.09 (±0.04)
NA-4	30	0.021 (±0.006)	0.79 (±0.14)
	40	0.029 (±0.004)	0.74 (±0.16)
	45	0.039 (±0.005)	0.55 (±0.18)
SA-1	30	0.041 (±0.004)	0.87(±0.13)
	40	0.040 (±0.010)	0.89 (±0.10)
	45	0.032 (±0.002)	1.13 (±0.19)
SA-2	30	0.011 (±0.004)	0.80 (±0.03)
	40	0.009 (±0.010)	0.91 (±0.04)
	45	0.011 (±0.002)	0.97 (±0.04)
SA-3	30	0.026 (±0.004)	1.16 (±0.16)
	40	0.034 (±0.010)	1.12 (±0.29)
	45	0.013 (±0.002)	1.34 (±0.17)
SA-4	30	0.005 (±0.004)	0.88 (±0.16)
	40	0.006 (±0.003)	0.78 (±0.31)
	45	0.008 (±0.001)	0.68 (±0.13)

^aThe reported error is characterized as 95% confidence intervals stemming from the repeatability of each measurement.

need to understand aerosol composition (or mineralogy in the case of dust) in order to accurately predict ClNO_2 yields as a function of RH. These results support our previous hypothesis regarding the importance of trace chloride-containing salts to facilitate the heterogeneous formation of ClNO_2 , even at a low RH. The consistently high yields observed with our playa samples, independent of the probed RH values (30–45%), can likely be explained by multiple processes including trace quantities of hygroscopic minerals coupled with the presence of hydrated salts—providing the aqueous chloride needed to produce ClNO_2 at a low RH.

The only sample which did not consistently produce high yields of ClNO_2 is NA-2. Instead, this sample showed increasing values of both $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 with an increase in RH. We speculate that aluminosilicates (commonly characterized by $^{76}\text{SiO}_3^-$ and/or $^{76}\text{AlO}_2(\text{OH})_2^-$ using SPMS)²⁸ in this sample are primarily reacting at low relative humidities, and, as RH increases, the saline evaporites are likely to deliquesce causing $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 to increase as shown in Table 1. Though we would expect $\gamma\text{N}_2\text{O}_5$ to decrease to ~ 0.03 and ϕClNO_2 to increase to ~ 1 as the salt begins to deliquesce and preferably react with N_2O_5 .²⁷ The increase in both $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 suggests that aluminosilicates and saline evaporites are heterogeneously distributed on the surface of the playa samples, rather than colocated, leading to both minerals reacting simultaneously and resulting in the increase in $\gamma\text{N}_2\text{O}_5$ with an increase in relative humidity.

Our observations are also consistent with a previous study quantifying the ϕClNO_2 of a single playa dust sample from the Great Salt Lake.^{27,28} Notably, sample SA-4 from this study is strikingly similar to the Great Salt Lake sample from Mitroo et al. (2019)²⁷ for ϕClNO_2 . Both samples were collected from

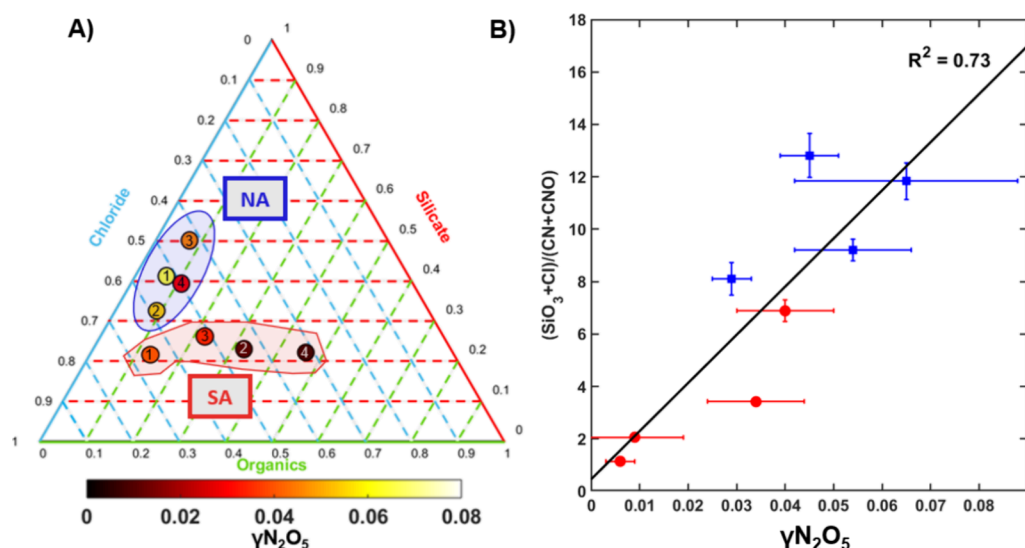


Figure 3. A) Ternary plot showing the SPMS intensity of average ion peaks for chloride ($^{35+37}\text{Cl}^-$), silicate ($^{76}\text{SiO}_3^-$), and organics ($^{26}\text{CN}^- + ^{42}\text{CNO}^-$) relative to one another. The individual points on this graph are shaded by the intensity of $\gamma\text{N}_2\text{O}_5$. B) Correlation between the ratio of silicate and chloride:organic species, $(^{76}\text{SiO}_3^- + ^{35,37}\text{Cl}^-):(^{26}\text{CN}^- + ^{42}\text{CNO}^-)$, versus $\gamma\text{N}_2\text{O}_5$. Playa samples from the Northern Area and Southern Area are blue and red, respectively. Error bars are characterized as 95% confidence intervals.

the same region of the lake (SA-4 is approximately 30 km northeast of the Mitroo et al. (2019) Great Salt Lake sample), and also share similar characteristic single particle mass spectra peaks including $^{79}\text{PO}_3^-$ and $^{56}\text{CaO}^+$.²⁸ Mitroo and colleagues tested a wider RH range for the Great Salt Lake sample (20–50%) and showed consistently high yields, similar to our results here. When we estimate ϕClNO_2 using the Bertram & Thornton (2009) parameterization,³¹ we also see a similar overestimate to Mitroo et al. (2019)²⁷ of the parameterized ϕClNO_2 compared to our observed ϕClNO_2 , which is discussed in detail in the SI.

3.3. Differing Reactive Uptake Coefficients of N_2O_5 between the Northern and Southern Areas of the Great Salt Lake. Playa dust generated from the Northern Area of the lake is, on average, twice as reactive with N_2O_5 (0.048 ± 0.013) as dust from the Southern Area of the lake (0.024 ± 0.016) (see Figure 2 and Table 1). To better understand potential factors driving the reduced reactivity in the Southern Area of the lake, we use the ternary plot shown in Figure 3A, which represents the relative ion intensities of organic nitrogen markers ($^{26}\text{CN}^-$ and $^{42}\text{CNO}^-$), silicates ($^{76}\text{SiO}_3^-$), and chloride ($^{35,37}\text{Cl}^-$) for single particles measured using SPMS. We use $^{26}\text{CN}^-$ and $^{42}\text{CNO}^-$ to represent organics as they are the most abundant organic peaks and represent nitrogen-containing organic matter in sediments and soil.^{28,108–111} As shown in Figure 3A, the reactivity of playas collected from the Southern Area of the lake are likely suppressed by the increased presence of organics, which prevents N_2O_5 from reacting with the dust but does not lower ϕClNO_2 .^{28,67,112–115} We suggest this could be due to the presence of organic material that does not fully coat the particle. The source of the organics in our playa samples is thought to be primary soil organic matter, rather than a secondary source, as vegetation was observed at several playa collection sites.⁷ The effect of organics are particularly noticeable in the Southern Area of the lake with samples SA-2 and SA-4, where $\gamma\text{N}_2\text{O}_5$ ranges from 0.005 ± 0.004 to 0.011 ± 0.004 and ϕClNO_2 remains relatively consistent ~ 0.8 . As the relative intensity of the organics decreases, however, the reactivity of N_2O_5 begins to

increase, which can be observed with samples SA-1 and SA-3 ($\gamma\text{N}_2\text{O}_5$ ranges from 0.013 ± 0.002 to 0.041 ± 0.006) in Figure 3A and Figure S8. Increased content of particulate silicates ($^{76}\text{SiO}_3^-$) and chloride ($^{35,37}\text{Cl}^-$) were observed in samples collected from the Northern Area of the Great Salt Lake as shown in Figure 3A and Figure S8. Figure 3B shows a strong positive correlation between the ratio of (silicates + chloride)/organics (i.e., $(^{76}\text{SiO}_3^- + ^{35,37}\text{Cl}^-):(^{26}\text{CN}^- + ^{42}\text{CNO}^-)$) versus $\gamma\text{N}_2\text{O}_5$ ($R^2 = 0.73$). Northern Area samples have the highest ratio of reactive species (silicate and chloride), suggesting that the increased reactivity of the Northern Area samples is due to an increased presence of silicates and chloride-containing evaporites and less organic aerosol than the Southern Area. Similar to ϕClNO_2 , $\gamma\text{N}_2\text{O}_5$ is overpredicted using the traditional N_2O_5 uptake parameterization,³¹ which is discussed in the SI, further demonstrating the importance of understanding aerosol composition when predicting $\gamma\text{N}_2\text{O}_5$.

We also determined how the different gypsum content between the Northern and Southern Areas of the lake, discussed in Section 3.1, affects our results. Gypsum is poorly soluble and has a high deliquescence RH (DRH > 99%).^{116–119} As such, we are unlikely to observe much interaction between N_2O_5 and $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ at the humidities probed in this study. As described in Figure 1, the Southern Area of the lake has a higher estimated concentration of gypsum, which we hypothesize is also reducing the N_2O_5 reactivity of Southern Area playa dust. To test our hypothesis, we added a gypsum standard into our aerosol flow system and observed a low reactive uptake ($\gamma\text{N}_2\text{O}_5$ ranges from 0.007 to 0.009, Figure S7), which, to the best of our knowledge, are the first measurements to quantify the reactive uptake of N_2O_5 to gypsum. By utilizing the predicted gypsum weight percent from ISORROPIA-II for our playa dust samples, we observe a negative correlation between predicted gypsum content and $\gamma\text{N}_2\text{O}_5$ ($R^2 = 0.29$, Figure S9) suggesting that the increased weight percent of CaSO_4 (assumed to be gypsum) in our samples suppresses the reactive uptake coefficients in the Southern Area of the lake in addition to organic matter. Previous observations of particulate sulfate and halide mixtures

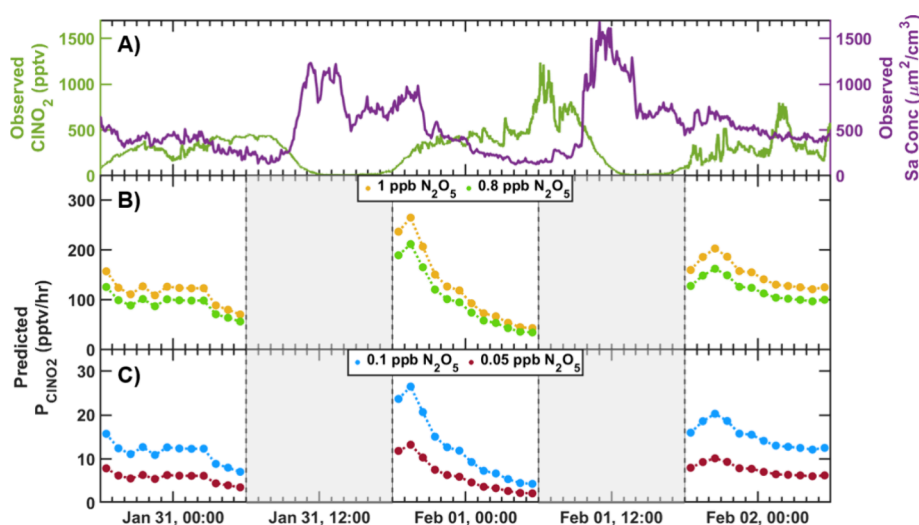


Figure 4. A) Observed concentration of ClNO_2 (green) and the estimated total coarse mode ($\text{PM}_{10} - \text{PM}_{2.5}$) surface area (Sa) concentration (purple) from January 30th, 2017, to February 2nd, 2017, during the UWFPS field campaign.²⁰ B) Hourly P_{ClNO_2} estimates from playa dust during noninversion conditions from 18:00 to 06:00 MST during the UWFPS campaign. C) Hourly P_{ClNO_2} estimates from playa dust during inversion conditions from 18:00 to 06:00 MST during the UWFPS campaign. Areas shaded out in gray in both B and C are approximately times when sunlight is present.

have shown a reduction in ϕClNO_2 when reacting with N_2O_5 , suggesting that sulfate can inhibit the production of ClNO_2 .¹²⁰ Given that ϕClNO_2 does not vary significantly between playa samples with higher particulate sulfate, we suggest that the sulfate ion in gypsum does not dissociate into the aqueous phase and affect halogen formation.

3.4. Estimating the Production of ClNO_2 from the Great Salt Lake Playa during the UWFPS Campaign. We next utilize our laboratory measurements to estimate the contribution of playa dust to ClNO_2 observations during the UWFPS campaign. The estimated nighttime production of ClNO_2 contributed by playa dust from January 30th, 2017 to February 2nd, 2017 is shown in Figure 4 and Table S6. Throughout the three nights of the PCAP, the estimated P_{ClNO_2} from playas ranges from 6 to 152 pptv/h when $[\text{N}_2\text{O}_5](t)$ is estimated to be 50, 100, 800, and 1000 pptv, and $\gamma\text{N}_2\text{O}_5 = 0.033$ and $\phi\text{ClNO}_2 = 0.88$ (which are the average $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 for all playa samples analyzed in this study). To further support that we are likely observing mineral dust, we compare soluble ion concentrations from our study to observations by Hrdina et al. (2021),¹⁸ which is described in Section 8 of the SI and shows that the ambient soluble ion concentrations match the chemical signature of evaporite minerals from playa dust. The first night (01.30.2017–01.31.2017) is calculated to produce the lowest estimated P_{ClNO_2} , which ranges from 6 to 114 pptv/h (averaged throughout the entire evening). This is because the coarse mode surface area concentration ($\text{PM}_{10} - \text{PM}_{2.5}$) is relatively low during this night, with an average surface area concentration of $379 \pm 14 \mu\text{m}^2/\text{cm}^3$. The second night (01.31.2017–02.01.2017) yielded a similar average P_{ClNO_2} , ranging from 6 to 123 pptv/h, which remains statistically indifferent from the first night for all four N_2O_5 estimates probed. The increased spread in these predictions is due to a large decrease in coarse mode surface area concentration after midnight on 02.01.2017, shown in Figure 4, leading to an increase in P_{ClNO_2} at the beginning of the evening, which decreases as the night progresses. The last night (02.01.2017–02.02.2017) yielded the largest P_{ClNO_2} , which is significantly

higher than the other two nights, and ranges from 8 to 152 pptv/h. This night was characterized by a high and relatively stable coarse mode surface area concentration, which persisted through the evening, leading to a relatively large and stable P_{ClNO_2} . We compared the estimated production of ClNO_2 from playa dust to ambient observations of ClNO_2 during the UWFPS campaign. Our P_{ClNO_2} estimates (from playa dust) account for 0.2 to 5.0% of ambient ClNO_2 observations during the PCAP event assuming $\text{Sa}(t)$ is equivalent between the UofU and Logan field sites.²⁰

Previous observations of PCAPs in Salt Lake City have observed injections of clean air into the basin, which can be characterized by a brief decrease in observed $\text{PM}_{2.5}$ over a few days, then a rapid increase in $\text{PM}_{2.5}$ above $60 \mu\text{g}/\text{m}^3$.¹⁰ A clean air injection can lead to the injection of mineral dust from the Great Salt Lake (and surrounding deserts and lake beds) into the cold air pool.¹⁰ However, a clean air injection was not observed during the UWFPS campaign to the best of our knowledge.²⁰ Since UWFPS likely did not observe a clean air injection, the total contribution of coarse mode mineral dust to surface area concentration during this study is likely an underestimate when compared to other years, which can result in a lower P_{ClNO_2} from mineral dust. Further, dust storms from the Great Salt Lake also tend to peak in the early spring (usually late March and early April).^{2,58,121} During this time, high concentrations of coarse mode particulate matter can be observed, likely in the form of playa dust from the Great Salt Lake and surrounding lake beds, which can react with N_2O_5 in the nighttime.² While these periods would likely yield a higher production of ClNO_2 , ambient observations of N_2O_5 , ClNO_2 , and coarse mode surface area concentration have not been measured during major dust storms. Further, our P_{ClNO_2} estimates neglect the contribution of fine mode dust aerosols ($\text{PM}_{2.5} - \text{PM}_{0.5}$) during the UWFPS campaign. Modeling studies and measurements including the effects of NO on ambient N_2O_5 concentrations and better apportionment of dust to both fine and coarse mode surface area concentration during PCAP events in Salt Lake City would likely improve (and increase) our estimates. Finally, we neglect contributions

from other nearby dust sources that lack reactive uptake and evaporite composition measurements, including the Sevier Dry Lake, Sevier Desert, and Great Salt Lake Desert that are also considered dust hotspots.^{2,9,122} To fully include playa dust as a halogen source in air quality models, similar lab and field studies with playas from Sevier Dry Lake, Sevier Desert, Great Salt Lake Desert, and other dust sources that affect Salt Lake City should be performed.

If P_{ClNO_2} is sustained over the course of an evening (18:00 to 06:00 MST), and we use the total estimated range of P_{ClNO_2} , we estimate that between 68 to 1819 pptv of ClNO_2 generated from playa dust-derived chloride could be produced throughout the night. When we compare our estimates to P_{ClNO_2} estimates in a polluted coastal region during the 2015 WINTER field campaign,⁵⁷ where ~ 1850 pptv of ClNO_2 is produced throughout the evening and sea spray aerosol is the primary source of particulate chloride, we see a nearly 1:1 comparison of nightly production of ClNO_2 between a polluted coastal site and playa dust in the Salt Lake Valley during the low-dust season. Even though the measurements of ClNO_2 between the UWFPs and WINTER field studies were different (ground-based measurements versus aloft aircraft measurements respectively), we find it important to highlight that the ClNO_2 producing ability of playa dust is comparable to that of a polluted coastal region where ClNO_2 has significant impacts on the oxidant budget, NO_x spatial distributions, and $\text{PM}_{2.5}$ formation. To reconcile ClNO_2 observations using computational models, especially in inland regions far from the ocean, nonsea-spray sources of particulate chloride (such as playa dust from the Great Salt Lake) need to be included as a source of ClNO_2 .

3.5. Atmospheric Implications. Our results demonstrate that playa dust from different regions of the same shrinking lakebed exhibit differing reactivity with N_2O_5 . In this case, dust from the Northern Area of the Great Salt Lake are, on average, twice as reactive with N_2O_5 when compared to playa dust sourced from the Southern Area of the Great Salt Lake. Differences in mineral composition are likely the primary control on playa dust reactivity, where high silicate and chloride content resulted in an increased $\gamma\text{N}_2\text{O}_5$, while elevated gypsum and organic content resulted in a reduced $\gamma\text{N}_2\text{O}_5$ in samples collected from the Southern Area. Despite these differences in reactivity, all of the playa dusts generated ClNO_2 at high yields ($\phi\text{ClNO}_2 > 0.5$), except for one sample, likely due to trace amounts of highly soluble potassium, calcium, and magnesium chlorides, which have previously been hypothesized to facilitate ClNO_2 production at low relative humidities.²⁸ We confirmed this mechanism can occur at low relative humidities by testing a MgCl_2 standard, which reacted efficiently with N_2O_5 and produced ClNO_2 at a low RH. We also confirm the low reactivity of gypsum with N_2O_5 , further underscoring the importance of mineralogy in controlling reactive uptake kinetics.

We extend our laboratory studies to estimate the contribution of playa dust sources to the production of ClNO_2 during the 2017 UWFPs campaign. The total production is estimated to be between 6 to 151 pptv/h (0.2 to 5.0% of the ambient ClNO_2 observed during a 3-day PCAP event), and dependent on the ambient surface area concentration of mineral dust and estimated concentration of N_2O_5 . Future field and modeling studies in Salt Lake Valley will likely find a larger contribution of ClNO_2 from playa dusts during the early spring, which is characterized by large dust

storms and emissions from the nearby lakes and deserts.^{2,9,58,122} Our results coupled with better source apportionment of playa hotspot sources in air quality models, will likely lead to a more robust prediction of poor air quality events in Salt Lake Valley from natural sources of particulate halides. Additionally, field studies utilizing single-particle measurements to quantify the contribution of playa dust to the total Salt Lake Valley aerosol throughout the year will likely prove to be an invaluable tool in achieving better source apportionment of chloride sources producing ClNO_2 .

When we compare our laboratory results to ambient parameterizations of ϕClNO_2 during the UWFPs field campaign,^{20,21} our average ϕClNO_2 for the playa data set ($\phi\text{ClNO}_2 = 0.88$) is approximately 4-times that of the UWFPs parameterization ($\phi\text{ClNO}_2 = 0.22$).¹²³ The higher yield from generated playa dust is not surprising as we are using “fresh” playa samples in a controlled laboratory setting rather than ambient wintertime polluted conditions to estimate ϕClNO_2 . When we compare our observations to other known ambient sources of ClNO_2 , such as the contribution of sea salt aerosol to ϕClNO_2 estimated during the 2015 WINTER field campaign ($\phi\text{ClNO}_2 = 0.138$),¹²⁴ we continue to see disproportionately high yields of ClNO_2 from playa dust. The disproportionately higher yields of ClNO_2 from playa dust highlights that playa dust aerosol can have a large impact on Cl radical production, and therefore air quality.

Our results highlight the need to better understand the future impacts of the shrinking Great Salt Lake (and other shrinking lakes) on halogen-initiated reactions, especially as reductions of anthropogenic halogen sources are implemented in the United States.^{55,125} Given the continued water level reductions in the Great Salt Lake, and brine lakes worldwide,^{1,3,56,126,127} the contribution of saline evaporite-rich dust to depreciating air quality will likely increase over time, which has already been observed with similar dried lakebeds.^{128–131} Previous laboratory studies have already begun to quantify the reactivity of similar dust sources.^{27,28,132} However, current air quality models do not factor in the production of reactive halogens from playa dusts.^{52,53} Further, studies using ambient techniques which can measure N_2O_5 reactivity and ClNO_2 formation^{39,79,133} in areas proximal to shrinking brine lakebeds or similar studies on playa dust reactivity^{27,28} are needed to better understand the impact of shrinking saline lakebeds on regional air quality.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsearthspacechem.4c00258>.

Background information regarding playa dust and water samples, detailed report on the chemical ionization mass spectrometer and single-particle aerosol mass spectrometer, diagram of the flow tube apparatus, validation of the particle modulation technique, corrections to surface area concentration, method used to determine soluble ion concentration, reactive uptake of N_2O_5 on mineral standards, comparison between laboratory observations and parameterized estimates of $\gamma\text{N}_2\text{O}_5$ and ϕClNO_2 , soluble ions from fine mode particulate matter and how they can act as indicators of playa dust during the UWFPs campaign, additional figures (PDF)

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Notes

The views expressed in this article are those of the authors and do not necessarily reflect the views or policies of the U.S. EPA. The authors declare no competing financial interest.

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