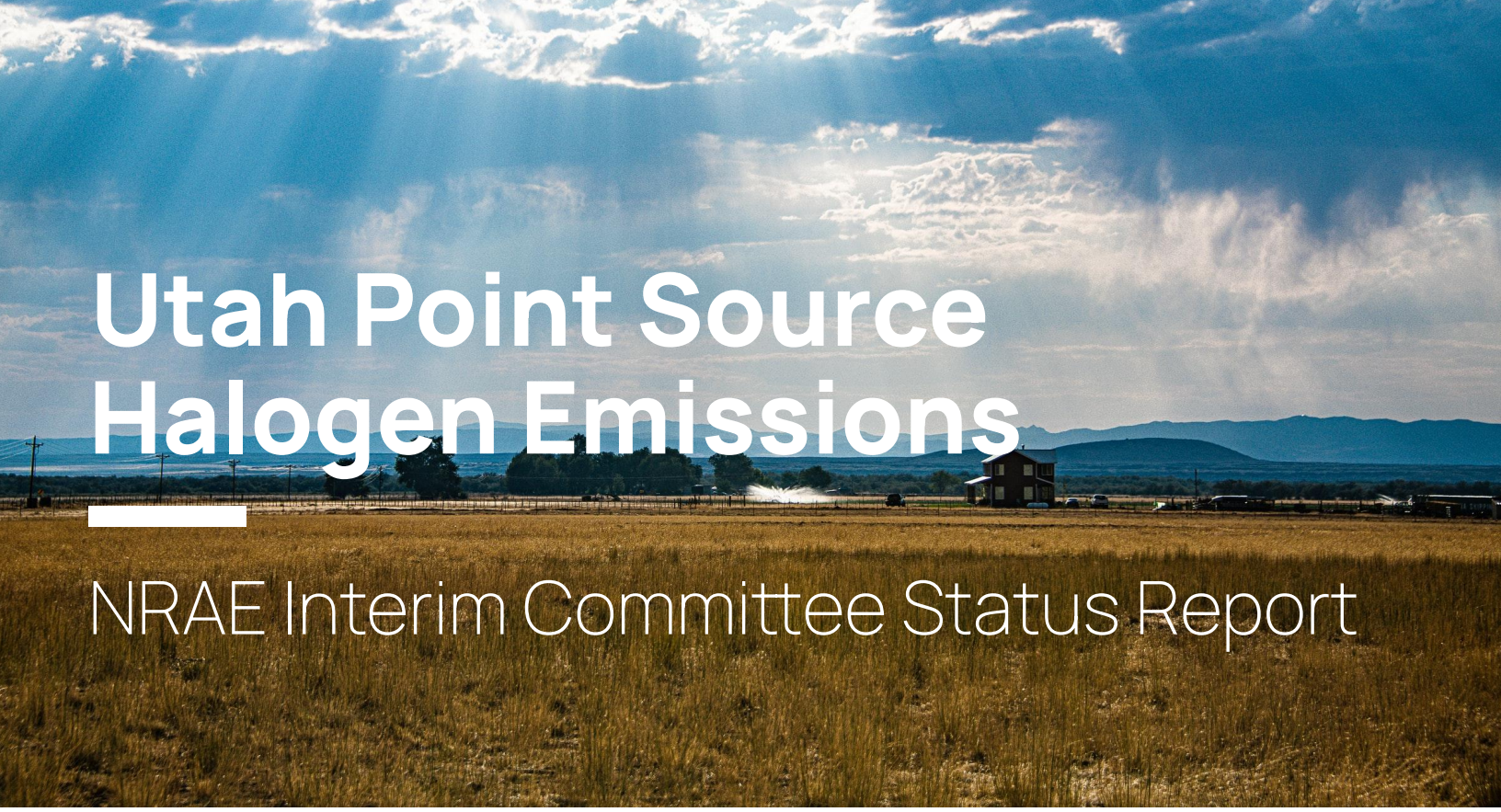

Utah Point Source Halogen Emissions

NRAE Interim Committee Status Report

PREPARED BY Utah Division of Air Quality

UTAH DEPARTMENT *of*
ENVIRONMENTAL QUALITY
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Background

HB 220 - Emissions Reduction Amendments, was passed during the 2023 Utah Legislature General Session. The legislation was subsequently signed into law by Governor Cox. The legislation's applicability is limited to Box Elder, Davis, Salt Lake, Weber, Tooele and Utah counties and directs the Utah Division of Air Quality (DAQ) to:

1. Complete an air emissions inventory of point sources that emit halogens by December 31, 2024.

2. Complete a best available control technology emissions reduction plan to reduce the compounds of halogens with an implementation date of December 31, 2026.

3. Provide recommendations for a state standard limiting halogen emissions.

These items are to be published on DAQ's website. The legislation also directs DAQ to report on the status of the above to the Natural Resources, Agriculture, and Environment (NRAE) Interim Committee during the November 2023 and 2024 meetings.

Current Halogen Information and Emission Inventory Status

Current Halogen Background

Key halogens such as chlorine, bromine, and iodine play an important role in atmospheric chemistry. They are reactive species that affect the oxidative capacity of the troposphere. The introduction of gaseous reactive halogen species, such as HCl, Cl_2, $ClNO_2$, Br_2, $BrNO_2$, and BrCl, leads to the production of oxidized Volatile Organic Compounds (VOCs), resulting in increased ozone (O_3) and fine particulate matter ($PM_{2.5}$) concentrations, especially under oxidant-limited conditions. Halogen sources include a mix of natural and anthropogenic sources[1,2], with anthropogenic sources mainly comprising area and point

[1] Fu, X.; Wang, T.; Wang, S.; Zhang, L.; Cai, S.; Xing, J.; Hao, J. Anthropogenic Emissions of Hydrogen Chloride and Fine Particulate Chloride in China. Environ. Sci. Technol. 2018, 52, 1644−1654.
[2] Simpson, W. R.; Brown, S. S.; Saiz-Lopez, A.; Thornton, J. A.; von Glasow, R. Tropospheric Halogen Chemistry: Sources, Cycling, and Impacts. Chem. Rev. 2015, 115 (10), 4035−4062.

sources. Fluorine is also considered a halogen but does not display the reactivity of the aforementioned key halogens and was thus not included in this analysis.

Sources of halogens in Utah and their emission rates are largely unknown, and the current emissions inventory for these compounds needs to be further resolved. An aircraft and ground-based field intensive study conducted by the National Oceanic and Atmospheric Administration (NOAA) along the Northern Wasatch Front (NWF) during a 2017 winter-time PM stagnation air pollution episode investigated the magnitude of industrial emissions of chlorine and bromine in Utah's airshed. Findings indicated that US Magnesium, an industrial source located on the Southwest edge of the Great Salt Lake, is a source of halogens in the Salt Lake Valley (SLV)[3]. Aircraft observations collected using an iodide chemical ionization mass spectrometer (I–CIMS) for bromine- and chlorine-containing species detection showed extreme levels of HCl and dihalogens Cl_2, Br_2, and BrCl in the plume originating from US Magnesium. These levels were also associated with complete ozone depletion in the vicinity of the plant but increased $PM_{2.5}$ formation downwind.

Modeling using these observed halogen emissions shows regional $PM_{2.5}$ increases of 10%–25%, with greater increases being observed closer to the plant. While these findings show an impact from halogens from US Magnesium on local $PM_{2.5}$ levels in the urban areas of the NWF, results are limited to a single winter campaign and limited plume encounters. Further measurements are needed to constrain Utah's emissions inventory for halogens and better understand the impact of these species on local $PM_{2.5}$ and ozone levels.

To better understand potential emission sources of halogens in Utah, a literature review of existing halogen emission inventories and field observations was conducted. The review showed that most studies have focused on reactive chlorine, with primary anthropogenic emission sources of reactive chlorine species (HCl, particulate Chloride (pCl), Cl_2, and HOCl) including power plants[4], industrial processes[5,6], wastewater treatment plants[7],

[3] C. C. Womack, W. S. Chace, S. Wang, M. Baasandorj, D. L. Fibiger, A. Franchin, L. Goldberger, C. Harkins, D. S. Jo, B. H. Lee, J. C. Lin, B. C. McDonald, E. E. McDuffie, A. M. Middlebrook, A. Moravek, J. G. Murphy, J. A. Neuman, J. A. Thornton, P. R. Veres, and S. S. Brown. Midlatitude Ozone Depletion and Air Quality Impacts from Industrial Halogen Emissions in the Great Salt Lake Basin. *Environ. Sci. Technol*. 2023, 57, 5, 1870–1881

[4] Riedel, T. P.; Wagner, N. L.; Dubé, W. P.; Middlebrook, A. M.; Young, C. J.; Ö ztürk, F.; Bahreini, R.; VandenBoer, T. C.; Wolfe, D. E.; Williams, E. J.; et al. Chlorine Activation within Urban or Power Plant Plumes: Vertically Resolved ClNO2 and Cl2 Measurements from a Tall Tower in a Polluted Continental Setting. J. Geophys. Res. Atmos. 2013, 118, 8702–8715.

[5] Fu, X.; Wang, T.; Wang, S.; Zhang, L.; Cai, S.; Xing, J.; Hao, J. Anthropogenic Emissions of Hydrogen Chloride and Fine Particulate Chloride in China. Environ. Sci. Technol. 2018, 52, 1644–1654.

[6] Liu, F.; Peng, M.; Dong, P.; Le, H.; Xu, X. Design and Analysis of Supplement Water and Supplement Water Storage Tank for Cooling Tower in Date Center. Constr. Sci. Technol. 2019, 19, 85–88.

[7] Mielke, L. H.; Furgeson, A.; Osthoff, H. D. Observation of ClNO2 in a Mid-Continental Urban Environment. Environ. Sci. Technol. 2011, 45, 8889–8896.

cooling towers[8], swimming pools[9], tap water usage, coal combustion[10,11], municipal solid waste incineration[12,13], biomass burning[14], cooking[15] and chlorine-containing disinfectants [16,17]. HCl is also released from sea-salt aerosol through acid displacement reactions[18].

Studies investigating anthropogenic bromine and iodine species are very limited. Li et al. (2021)[19] developed a bottom-up emission inventory of reactive bromine species from coal combustion activities, including power plants, industrial processes (e.g, cement, iron and steel, brick, lime production), industrial boiler, and residential burning, in China. After thorough review of the literature and consultation with scientific experts, including the Environmental Protection Agency (EPA) and NOAA, DAQ narrowed down the list of halogens species with potential primary emissions to dihalogens (Cl_2, Br_2, I_2, BrCl, ICl, IBr) and halogen halides (HCl, HBr, HI).

While fluorine is a halogen, fluorine compounds are not considered since they are not expected to influence tropospheric ozone formation. Fluorine is typically converted into HF, which is highly unreactive[20].

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[8] Chang, S.; Allen, D. T. Atmospheric Chlorine Chemistry in Southeast Texas: Impacts on Ozone Formation and Control. Environ. Sci. Technol. 2006, 40, 251–262.
[9] Faxon, C. B.; Allen, D. T. Chlorine Chemistry in Urban Atmospheres: A Review. Environ. Chem. 2013, 10, 221–233.
[10] Fu, X.; Wang, T.; Wang, S.; Zhang, L.; Cai, S.; Xing, J.; Hao, J. Anthropogenic Emissions of Hydrogen Chloride and Fine Particulate Chloride in China. Environ. Sci. Technol. 2018, 52, 1644–1654
[11] Liu, Y.; Fan, Q.; Chen, X.; Zhao, J.; Ling, Z.; Hong, Y.; Li, W.; Chen, X.; Wang, M.; Wei, X. Modeling the Impact of Chlorine Emissions from Coal Combustion and Prescribed Waste Incineration on Tropospheric Ozone Formation in China. Atmos. Chem. Phys. 2018, 18, 2709–2724.
[12] Finley, B. D.; Saltzman, E. S. Measurement of Cl2 in coastal urban air. Geophys. Res. Lett. 2006, 33, L11809.
[13] McCulloch, A.; Aucott, M. L.; Benkovitz, C. M.; Graedel, T. E.; Kleiman, G.; Midgley, P. M.; Li, Y.-F. Global Emissions of Hydrogen Chloride and Chloromethane from Coal Combustion, Incineration and Industrial Activities: Reactive Chlorine Emissions Inventory. J. Geophys. Res. Atmos. 1999, 104, 8391–8403.
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[15] Qiu, X.; Ying, Q.; Wang, S.; Duan, L.; Zhao, J.; Xing, J.; Ding, D.; Sun, Y.; Liu, B.; Shi, A.; et al. Modeling the Impact of Heterogeneous Reactions of Chlorine on Summertime Nitrate Formation in Beijing, China. Atmos. Chem. Phys. 2019, 19, 6737– 6747.
[16] Li, L.; Yin, S.; Huang, L.; Yi, X.; Wang, Y.; Zhang, K.; Ooi, C. G.; Allen, D. T. An Emission Inventory for Cl2 and HOCl in Shanghai, 2017. Atmos. Environ. 2020, 223, 117220
[17] Chang, S.; Allen, D. T. Atmospheric Chlorine Chemistry in Southeast Texas: Impacts on Ozone Formation and Control. Environ. Sci. Technol. 2006, 40, 251–262.
[18] Kelly, J. T.; Bhave, P. V.; Nolte, C. G.; Shankar, U.; Foley, K. M. Simulating Emission and Chemical Evolution of Coarse Sea-Salt Particles in the Community Multiscale
[19] Q. Li, X. Fu, X. Peng, W. Wang, A. Badia, R. P. Fernandez, C. A. Cuevas, Y. Mu, J. Chen, J. L. Jimenez, T. Wang, A. Saiz-Lopez. Halogens Enhance Haze Pollution in China. *Environ. Sci. Technol.* 2021, 55, 20, 13625–13637.
[20] Treatise on Geochemistry, Volume 4, 2003, 1-67. Chapter 4.02 Tropospheric Halogen Chemistry. R. von Glasow and P. J. Crutzen

Current Emissions Inventory

In order to develop the inventory of point sources that emit halogens, there first was a need to decide which facilities would be a part of the inventory based on whether they were already reporting halogen emissions or if they were potentially emitting halogens and not reporting. Point sources submit emissions inventories to DAQ's online State and Local Emissions Inventory System (SLEIS) database. Based on the information in the database, point sources already reporting halogen species were added to the halogen inventory after filtering for the counties in the nonattainment area and the key halogens of concern outlined by DAQ staff.

To account for facilities potentially emitting unreported halogens, staff reviewed available research on the types of facilities and processes known to emit halogens. Using the types of sources indicated in the research, a second list of potential facilities for the halogen inventory was created, filtering for the counties in the nonattainment area. The table below shows the list of potential sources of halogen emissions:

Table 1. Potential Halogen Emitting Sources.

Source	Potential Halogens	Notes	Included in Point Source Inventory search?
Cooling towers (multiple facilities)	HOCl, Cl_2	Based on Chang study, the source is from biocides used in the tower. Yin et al. ACS Earth Space Chem. 2022, 6, 1846−1857	Included through Source Category Code (SCC) Description, Emission Unit Description, and Process Description
Waste incineration	HCl, pCl	Yin et al. ACS Earth Space Chem. 2022, 6, 1846−1857	Included through the SCC Description, Emission Unit Description, and Process Description
Coal combustion (subsectors power plants, industrial boilers, kilns..)	Cl_2, HCl, pCl BrCl, HOBr, Br_2	Yin et al. ACS Earth Space Chem. 2022, 6, 1846−1857 Li et al. ES&T 2021	Included through Throughput Material "Coal Throughput"
Industrial processes (cement production, lime production, iron-steel production, brick production, glass production, chemical production, pickling industry)	HCl, pCl	Yin et al. ACS Earth Space Chem. 2022, 6, 1846−1857	Included by selected NAICS codes. No results for glass manufacturing
Biomass burning	HCl, pCl	Yin et al. ACS Earth Space Chem. 2022, 6, 1846−1857	No results for NAICS code 221117 or throughput material of "Wood Waste" or "Wood/Bark"

Source	Potential Halogens	Notes	Included in Point Source Inventory search?
Chlorine-containing disinfectants/Sewage plant (domestic, from hospitals)	HOCl, Cl_2	Yin et al. ACS Earth Space Chem. 2022, 6, 1846−1857	Included NAICS code 221320.
Power plants (coal, natural gas, diesel, fuel oil)	HCl, Br_2, BrCl, Cl2, pCl Iodine Potentially HBr (according to Sarwar Golam at EPA)	Lee et al. JGR 2018 + USGS fact sheet 2012 Wu et al. ES&T 2014	Yes (same NAICS code as coal combustion)
Aerospace		DAQ Group Discussion	Included through ATK facilities, Throughput Materials "Hydrogen" and "Solid Propellant"

Although there is some data on halogen emissions in the SLEIS database, there are gaps in the information. Many halogenated chemicals, including those with bromine, are not registered with the EPA as Hazardous Air Pollutants (HAPs) and are not required to be reported in emission inventories. There is also a lack of available emission factors for most halogens.

There are few studies published that discuss halogen emissions, one of which includes a study by Chang et al. at the University of Texas. This study examines possible sources for atomic chlorine precursors in Southeast Texas, including cooling towers, wastewater treatment facilities, and other area sources.

Cooling towers utilize chlorine or other chemical additives to control biofouling, the accumulation of organic microorganisms (such as the legionella bacterium responsible for Legionnaires' disease) on the inside surface of the towers. This addition of biofouling control agents can lead to a flash-off of chlorine species, where the air stream flowing through the cooling tower strips volatile chlorine (or other chemicals) constituents from the cooling water. Flash-off is the leading cause of chlorine emissions from cooling towers.

In order to estimate the amount of chlorine released by flash-off, there needs to be specific data collected from facilities about the use of chlorinated additives in cooling towers and the throughput and use of the cooling towers. This information is not readily available from point sources, and therefore, DAQ cannot use the emission factors in this study to apply to facilities with cooling towers. However, due to the small amount of chlorine additive in cooling towers, discontinuation of using chlorine in favor of bromine, and control devices such as drift elimination, DAQ staff concluded that chlorine emissions from

cooling towers are negligible. Chang et al. also came to the same conclusion when investigating wastewater treatment facilities.

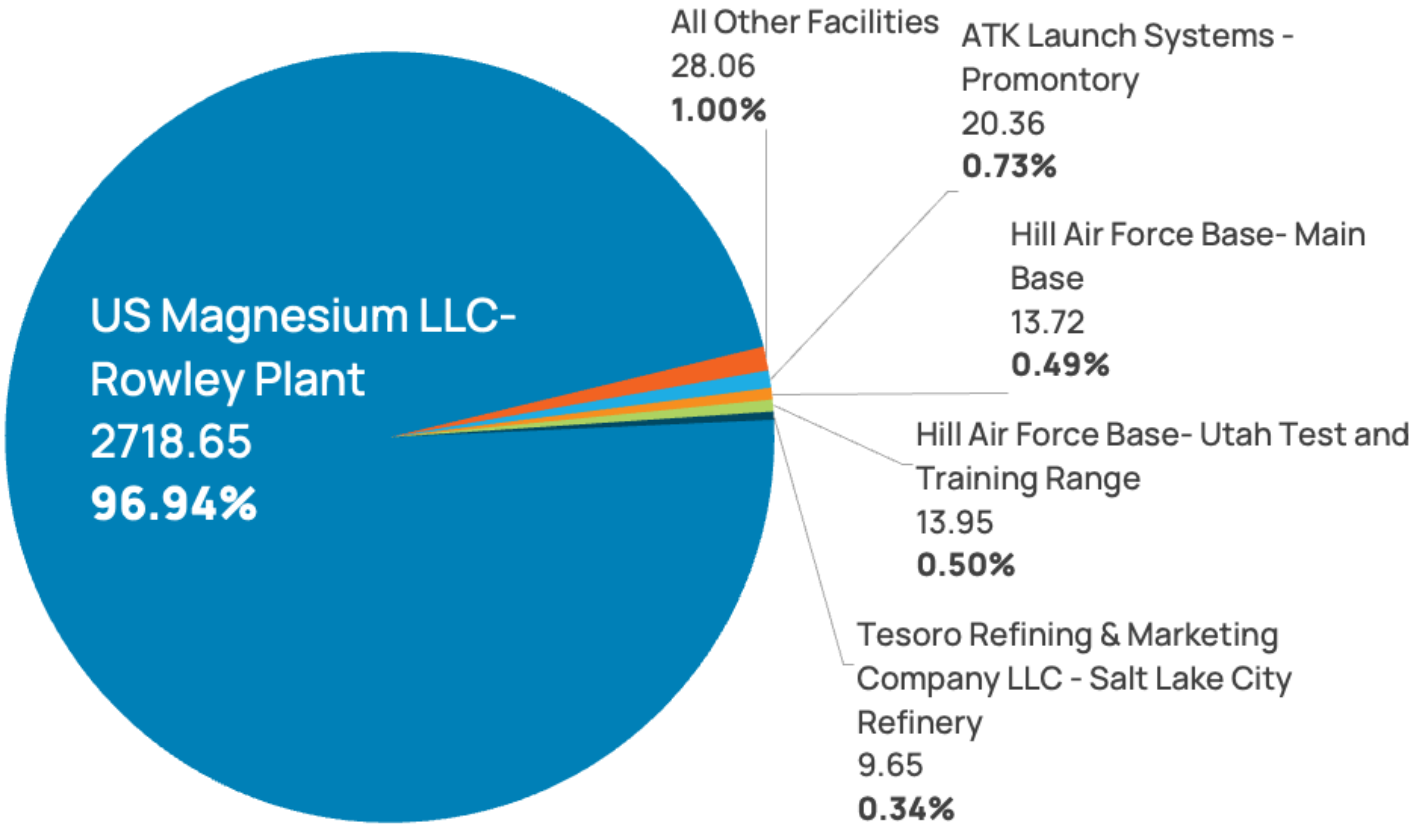
While the current DAQ point emissions inventory does contain several chlorine compounds, eight reactive chlorine gas compounds have been reported to constitute the majority of the tropospheric chlorine burden.[21] These compounds include: methyl chloride (chloromethane), hydrochloric acid, methyl chloroform (1,1,1-trichloroethane), methylene chloride, chloroform, Cl2, phosgene and chlorodifluoromethane.

Based on the above findings, current DAQ point source emissions inventory data (2020-2022) was compiled into three data sets available in the appendix and which will be posted to the DAQ website after NRAE committee review. The first data set, "Potential Halogen Point Source Inventory", is composed of all point sources that have the potential to emit a halogen compound based on the literature review and internal discussion (Listed in Table 1). The second data set, "Halogen Specific Point Source Inventory", is composed of currently available point source emissions which include any halogenated compound regardless of reactivity. The third data set, "Eight Reactive Chlorine Compound Emissions by Facility", is composed of point source emissions for the eight reactive chlorine gas compounds identified by Burklin et al. (note: only six of the eight compounds are currently found in the emissions inventory; phosgene and chlorodifluoromethane were not reported by any point sources). Figure 1 below provides a summary of the point source facilities emissions of the reactive chlorine gas compounds.

As was previously stated, the inventory is primarily limited to chlorinated compounds and few brominated and iodine-containing compounds. This again highlights the gaps related to non-chlorine and non-HAP halogen compounds and the need for entirely new emission factors for these types of compounds before any additional emissions could be calculated and added to an inventory. New emission factor development typically takes several years to complete, if determined to be feasible. The identification of potential sources and development of new emission factors is discussed in the stack testing and research sections below.

[21] Burklin, Clinton & Erickson, Steve & Bursey, Joan. (2023). Development of an Area and Mobile Sources Inventory for Reactive Chlorine Compounds in Southeast Texas.

Figure 1. Eight Reactive Chlorine Gas Compound Emissions From Point Sources (2020-2022 Point Source Inventory in tons/year)



Finally, Chang et al. also noted emissions from swimming pool water treatment. While not a point source, DAQ staff also evaluated the potential to estimate emission from these sources and determined that we currently do not have the activity data (population of pools or hot tubs in each county) and these emissions would also likely be negligible for Utah given the difference in number of pools compared to large metropolitan areas in southeast Texas (where the Chang et al. study took place).

Current And Future Halogen Research

Stack Testing

Stack testing consists of two parts: sample collection from the stack and the analytical method used to analyze the collected sample. US Magnesium is already required to test for

chlorine (Cl_2) and hydrochloric acid (HCl) using EPA Reference Method (RM) 26A. RM 26A can also be used to measure bromine (Br_2), hydrobromic acid (HBr), and hydrogen fluoride (HF). RMs 13, 13A, and 13B can be used to sample for fluorides (measured as fluorine). There are no other EPA Reference Methods for stack testing the other halogens.

To perform stack testing for the other halogens, such as iodine, DAQ would have to work with the sources and an analytical lab to develop appropriate stack testing procedures. Engineering estimates of emissions of the other halogens would have to be made available to ensure that any stack testing would meet detection limits and provide representative data before any other halogen stack test methods could be developed/implemented. Any halogen stack tests not currently required by permit would have to be ordered by the DAQ Director in consultation with the Utah Attorney General's Office.

US Magnesium's air quality permit requires them to perform RM 26A stack tests on their 05/06 Magnesium Chloride Bins, Spray Dryers 01-03, Melt Reactor, Emergency Off-gas Stack, and Hydrogen Chloride Production Unit. These units are currently not operating, however the Division of Air Quality has issued an order (DAQC-932-23, dated August 31, 2023) requiring all of these units to use RM26A to measure HCl, Cl_2, Br_2, and HBr within 180 days of achieving 90% of maximum magnesium production which is anticipated to occur in 2024.

Ongoing Halogen Measurement Studies

In collaboration with Utah State University, DAQ is currently conducting a three-phase sampling study to better understand the levels, spatial distribution and temporal variation of select halogens (HCl, HBr, pCl, pBr) along Utah's Wasatch Front. Measurement locations include 15 sampling sites distributed around the Great Salt Lake and along the Wasatch Front urban corridor, with sampling occurring over three sampling phases.

The first two sampling phases of the study, winter 2022-2023 and summer 2023, which corresponded to periods during which the magnesium production operation of US Magnesium was offline, have been completed, and the third phase is scheduled for winter 2023-2024. Concurrent seasonal ambient measurements of bromine oxide (BrO), nitrogen dioxide (NO_2) radicals and formaldehyde (HCHO) are also being collected by the University of Colorado at one of the 15 sampling stations. When complete, these studies will help constrain the spatial variability of select halogens, identify potential hotspots along the Wasatch Front and better model the impacts of halogens on air quality along the NWF.

Upcoming Summertime 2024 Field Campaigns

To determine factors and key precursor chemical species leading to ozone exceedances along the NWF, NOAA is planning a field measurement campaign in summer 2024. Through funding from DAQ, observations will be collected using a combination of aircraft, mobile (i.e. instrumented vehicle) and fixed ground-site measurements. Collected measurements will help determine the spatial distributions, speciation and sources of ozone precursors along the NWF. The role of halogens (chlorine, bromine and potentially iodine compounds), their sources (both industrial and natural sources, such as the Great Salt Lake) and their impact on ozone formation will specifically be investigated.

These measurements will also be augmented by measurements planned by other researchers for the same time period and region. Collaborators from Colorado State University will be collecting supplemental aircraft measurements using the National Science Foundation's King Air, and the University of Wyoming will be collecting additional mobile measurements using their mobile laboratory. These joint efforts will help maximize our understanding of factors leading to O_3 formation along the Wasatch Front and will help refine our emissions inventory, including our halogens source inventory.

The culmination of these efforts is anticipated to allow DAQ to identify additional sources of halogens and consider additional stack testing and/or emissions testing. These will provide the necessary steps to develop emissions factors to reduce 'halogen gaps' in future inventories and to address HB220 requirements related to control technologies and reduction plans.

Best Available Control Technology Emissions Reduction Plan Status

A Best Available Control Technology (BACT) analysis is case-by-case evaluation to determine an emission limit or operational control through the evaluation of technical feasibility, energy considerations, environmental impacts, and economic factors. [40 CFR 52.21(b)(12)]

A BACT analysis is performed for pre-construction permits for new or modified sources. A simplified explanation for BACT is the comparison of the proposed potential to emit (PTE) for the facility to the cost of reducing that PTE with a control or operational practice. Proposed controls can be eliminated through the BACT process if they are considered

technically infeasible within an industry and/or cost excessive in regards to dollars per ton of pollutant removed.

BACT analyses are submitted and created by facilities to outline applicable controls measures and operational limitations. BACT analyses are conducted by the source (or consultants hired by the source) as they are the experts on the individual industrial processes present at a given facility. Submissions include manufacture quotes, capital costs, and, when applicable, construction/installation costs.

Each halogen-emitting point source identified in this review is a permitted facility. As such, each operation was evaluated and required to submit a BACT analysis for criteria pollutants and HAPs prior to construction and installation of the applicable equipment in their Notice of Intent (NOI). Thus, a BACT evaluation was already performed on these operations.

Since the installation, it is possible that additional controls have arisen or become technically or economically feasible in the applicable industries. However, there is no regulatory precedent to request and implement a new BACT analysis or non-regulatorily justified controls for existing sources.

While there are many small, distributed sources of halogen emissions as identified in the literature and inventory described above, emission reduction strategies for these types of area sources tend to focus on one of two strategies: 1) the implementation of best practices to reduce emissions associated with loss, or 2) the development and implementation of alternative productives that have lower concentrations of the targeted pollutant. At this time, neither of these strategies for reducing distributed halogen emissions appear likely to be feasible at a large scale due to the lack of alternative products or due to the relatively small amount of emissions. Furthermore, public health considerations must also be taken into account for some of these sources (such as cooling tower treatment to mitigate Legionnaires' disease risk).

For large, major point sources of emissions, the installation of emission reduction control technologies might be a viable strategy for reducing primary emissions of halogens. As demonstrated in Figure 1, a single point source in the studied counties is responsible for 97.92% of the total point source emissions of halogens. Emission reduction strategies for large point sources are typically scrubbers, either chemically or thermally driven, but are not limited to these technologies.

There could be a benefit to requesting a halogen emitting source to perform a BACT, or BACT-like, analysis, but it is not currently within the authority of DAQ. If a BACT-like

evaluation were required by a special request, the legislature would need to define the parameters for review and control selection. Each facility would need to evaluate their existing process, determine applicable and available controls, seek manufacturing quotes and construction costs, and submit that information to DAQ for evaluation. There must be a reasonable cost per ton of emission reduced threshold for DAQ to select appropriate controls.

Halogen Standards Recommendation Status

Creating a state standard to limit or "cap" halogen emissions would be a considerable step as DAQ does not currently limit pollutants outside the Criteria pollutants subject to the National Ambient Air Quality Standards, as defined in the Clean Air Act,[22] at the state level. Similarly, the EPA does not limit most halogen emissions at the federal level. State Rules could be evaluated to propose and implement best practices, control technologies, or operating procedures, however, due to the variety of sources, it is unlikely that any one rule would be suitable for the variety of facilities. Rather, pollutants are limited on a permit-by-permit basis when facilities apply for their initial air permit or modifications to their existing permit. Each permit lists a Summary of Emissions table showing "an estimate of the total potential emissions from the source." All pollutants evaluated by DAQ's permitting group are listed, but staff only consider Criteria and Hazardous Air Pollutants in the evaluation. Staff do not evaluate halogens that do not fall into these two categories. DAQ uses these estimates to assign facilities into different categories, which bring additional requirements to the facility's permits. DAQ's permitting group may also list specific conditions for limiting individual pollutants based on guidance from EPA and the requirements of the Clean Air Act.

DAQ recommends continuing the established practice described above for limiting pollutants on a permit-by-permit basis for all Criteria and Hazardous Air Pollutants. For halogens not in these two categories, DAQ recommends asking for EPA guidance to standardize any limitations with other state, local, and tribal entities. As additional information is derived from the aforementioned studies and subsequent studies, DAQ will evaluate developing new rules, within the existing constraints of Utah Code Title 19, governing halogen emissions.

[22] 42 U.S.C. § 7409; see also implementing regulations at 40 C.F.R. 50.1 et seq.

Appendix

Potential Halogen Point Source Inventory

Halogen Specific Point Source Inventory

Eight Reactive Chlorine Compound Emissions by Facility