ENVIRONMENTAL RESEARCH
LETTERS

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Widespread impacts to precipitation of the East Palestine Ohio train accident

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Keywords: atmospheric wet deposition, high pH, fire impact, precipitation chemistry, back-trajectory (HYSPLIT) modeling, NADP, chloride

Supplementary material for this article is available online

OPEN ACCESS

RECEIVED
20 December 2023

REVISED
27 April 2024

ACCEPTED FOR PUBLICATION
31 May 2024

PUBLISHED
19 June 2024

Abstract

On 3 February 2023, a Norfolk Southern train derailment occurred in East Palestine, Ohio. The accident and subsequent fire resulted in the emissions of large amounts of hazardous compounds to the ambient atmosphere over many days. We used precipitation chemistry measurements routinely collected by the National Atmospheric Deposition Program (NADP) to estimate the spatial extent and chemical compounds deposited as a result of the accident. Our measurements revealed a large areal impact from the Midwest through the Northeast and likely Canada, and perhaps as far south as North Carolina (portions of 16 states, 1.4 million km^2). Observations showed the expected high chloride concentrations, but also unexpectedly high pH (basic) and exceptionally elevated levels of base cations exceeding 99th percentiles versus the historic record. These results were consistent with the meteorological conditions and atmospheric trajectories, and were not due to highly-concentrated low volume precipitation samples or wildfires. The robust measurements of the NADP network clearly show that the impacts of the fire were larger in scale and scope than the initial predictions, and likely due to the uplift from the fire itself entraining pollutants into the atmosphere. A more detailed evaluation of the accident and resulting fire could further refine the full impact of the atmospheric concentrations, dry and wet deposition, and the more specific extent of the spatial impact.

1. Introduction

At approximately 9 p.m. on the evening of 3 February 2023, a Norfolk Southern freight train accident occurred in East Palestine Ohio, less than one mile from the Pennsylvania border. About 50 train cars were involved in the accident, of which 38 derailed and at least 11 cars were carrying a variety of hazardous materials. Some of these materials were spilled, and a fire ensued. The subsequent fire burned for a number of days and damaged other cars [1, 2].

The cargo carried by the involved railroad cars included several volatile organic compounds (VOCs), including vinyl chloride, 2-butoxyethanol, 2-ethylhexyl acrylate, etc. As the fire continued, authorities attempted to control the fire and the flow of contaminated water, and, as reported, used a small amount of firefighting foams at the scene to control the fire. On 6 February, fire officials conducted what was described as a 'controlled release and burn' of five cars containing vinyl chloride (specifically vinyl chloride monomer, VCM), due to fear of an explosion of one or all of these cars. These cars were breached, the liquid was drained into a trench dug at the scene, and the VCM were set afire and burned. The evacuation order for the surrounding communities was lifted on 8 February, bringing an end to the active fire incident [1].

There were many reports of contaminated water and strong odors during and after the fire. The US Environmental Protection Agency (EPA) began atmospheric monitoring for a number VOCs in and around East Palestine soon after the accident. Significant atmospheric concentrations of many

Table 1. Summary of train cars involved in the accident, their cargo/content, and likely extent of combustion. Information summarized from currently available EPA tables and news reports [1, 2]. Cars with no known emissions are omitted.

No. of cars	Car type	Reported commodity	Extent of Comb.	Estimated Car volume	Notes
5	TANK	Vinyl Chloride (stabilized)	Complete	Combined to 422,380 l or so.	UN1086, cars subject to controlled explosion
4	TANK	Petroleum Lube Oil	Partial to complete	Entire load; 130,600 l max.	Assumed to be 80%–90% petroleum hydrocarbon distillates and 10%–20% additives
2	HOPPER	Polyethylene	Complete	76.6–151 m^3	Assumed to be solid beads
2	HOPPER	Polyvinyl	Complete	76.6–151 m^3	Assumed to be Polyvinyl Chloride (PVC) powder
3	TANK	Polypropyl, and Propylene Glycol	Near complete	Most of 130,600 l max.	
2	TANK	Diethylene Glycol	Complete	Entire load; 130,600 l max.	
2	HOPPER	Powder Flakes	Complete	76.6–151 m^3	Unclear what was in this car
1 each	TANK	EthylHexyl Acrylate, Butyl Acrylates (stabilized), and Isobutylene	Near complete	130,600 l max.	
1	TANK	Fuel Additives	Partial	130,600 l max.	Unclear what was in this car
1	TANK	Paraffin Wax	Partial	Unclear; 130,600 l max.	
1	HOPPER	Semolina	Complete	76.6–151 m^3	Generally coarsely milled durum wheat, but also other varieties or grains (rice, corn).
1 each	BOX	Medical Cotton Balls, Sheet Steel, frozen Vegetables	Complete	Total loss; 140–215 m^3	

chemicals were measured [3], including relatively high concentrations ($>1\ \mu g\ m^{-3}$) of acetone (consistently high), benzene, and other VOCs, and somewhat lower concentrations ($>0.1\ \mu g\ m^{-3}$) of ethylbenzene, naphthalene, tetrachloroethene, toluene, and p-xylenes. Table 1 contains a summary of the train's derailed cargo [2].

Clearly, for multiple-days, hazardous-classified chemicals were released into the atmosphere, both from spilled cargo and from the ensuing fire. However, much of the accident's impact upon the surrounding population and environment requires further study, including a complete accounting of the emissions and areal impact. Initial reports predicted the combustion products of VCM [hydrochloric acid, carbon monoxide and phosgene gas ($COCl_2$)], along with a number of other VOC emissions. Several recent articles in the published and grey literature describe aspects of the emissions [4, 5].

The US federal government, states, tribal organizations, and other researchers and collaborators operate the National Atmospheric Deposition Program (NADP) [6], a suite of integrated networks designed specifically to measure the wet deposition of atmospheric pollutants in precipitation into the North American biosphere. The NADP-NTN (National Trends Network) collects weekly precipitation samples at approximately 260 sites across North America, and quantifies levels and deposition of acid anions, base cations and several other analytes. Using

these measurements, we examined the precipitation chemistry and wet deposition of precipitation acidity and selected inorganic constituents for indication of the accident's areal impact on atmospheric chemistry (figure S1). The NADP networks were successfully used previously for this type of investigation [7].

2. Brief methods and materials

The NADP-NTN provides a long-term record (since 1978) of precipitation chemistry at generally rural sites in North America. Each site makes a weekly collection of precipitation (Tuesday to Tuesday) with a wet-only precipitation collector and precipitation gage. The automated collector opens during precipitation events (rain and snow), and closes when no precipitation is occurring. Site operators follow standard operating procedures and collection times, and the laboratory has strict quality assurance for accurate and representative measurements. A multistep validation for every sample occurs following the documented NTN protocols (see [6] for additional information).

All samples are analyzed for free acidity (H^+ as pH), specific conductance, calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), sulfate (SO_4^{2-}), nitrate (NO_3^-), chloride (Cl^-), and ammonium (NH_4^+) ions. NADP standard instrumentation was used for the analysis, and annual detection limits, blank and calibration results are all noted for each analyte in our annual quality assurance reports [6]. Detection limits for each analyte were below 0.01 mg l^{-1} (<0.01 pH units). For each analyte the concentration was measured (mg l^{-1} in precipitation), and the wet deposition (or flux, mg ha^{-1} week) determined using the rate of precipitation (mm $week^{-1}$) [6].

Data were used from all NTN sites in the Northeast, Midwest and Mid-Atlantic regions that had measurable precipitation and samples during the accident period (approximately 65 sites, figure S1).

We compared the NTN wet deposition measurements from the week of the train accident (31 January–7 February), and the NTN collection week immediately after the accident (7 February–14 February) to the previous 11 years of wintertime observations for the same sites and compounds. For this analysis, data from 9,800 samples were used, with 9,049 being valid samples for pH and other analytes. Of these, 8,720 samples were from the historic period prior to the accident, 56 samples were from the accident week, and 51 samples were available for the week after the accident. Many of these accident week samples were flagged for contamination (a total of 30 samples in Pennsylvania, Michigan, Massachusetts, Wisconsin, and almost every site in New York), with many flagged by the operator as having 'soot/ash/dirt' in the samples. This contamination designation is not uncommon for rural NTN sites, but in this situation is indicative of impact directly from the accident. During the accident week, much of Pennsylvania was dry and therefore did not have a wet sample.

Basic statistics and graphical results were generated using SAS software [8]. Estimates of atmospheric transport were made using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model with its forward and backward trajectory functions and Global Data Assimilation System meteorological data [9].

3. Observations and discussion

3.1. Atmospheric conditions

During the two-week period from just before to after the accident, widespread precipitation, most in the form of snow, occurred in the upper Midwest and Northeast (figures 1 and S2) [10]. However, much of the region from Illinois to Maryland was dry, including many areas in Pennsylvania and Ohio, and therefore this area had limited wet deposition collections. Had precipitation occurred here, more observations of the wet deposition impact would have been available.

On the morning of the accident (3 February), a cold front pushed through the accident region, with surface air flowing towards the south, and upper air (500 mb level) winds to the northeast. Light precipitation was occurring along the Canadian/New York border, which continued most of the week. On 4 February, a stationary front was situated to the northeast of East Palestine, with warmer air flowing north over the area and light precipitation again occurring along the border. Upper airflow remained towards the east. On 5 February, the stationary front had cleared out of the area, surface and upper level winds were to the northeast, and precipitation was occurring in southern Ontario through western Maine. A cold front was approaching from the north. On 6 February, (the day of the VCM breach), surface high pressure was situated to the west and low pressure to the east, both of which resulted in southerly surface flow. The upper level situation caused air to move into Virginia and then back to the Northeast. Light precipitation was again occurring along the Canadian/New York border to western Maine, with little other precipitation in the Midwest and Northeast.

On the morning of 7 February, an area of low pressure was situated over Lake Michigan, with a low pressure trough aloft, and an occluding cold front at the surface. Surface air at the accident site was flowing north, with significant precipitation occurring from southwest Maine to Minnesota, and heavier precipitation in southern Canada. With cyclonic rotation around this low pressure, winds were moving from the accident region to the north and wrapping around into Michigan, Wisconsin, and Canada [11].

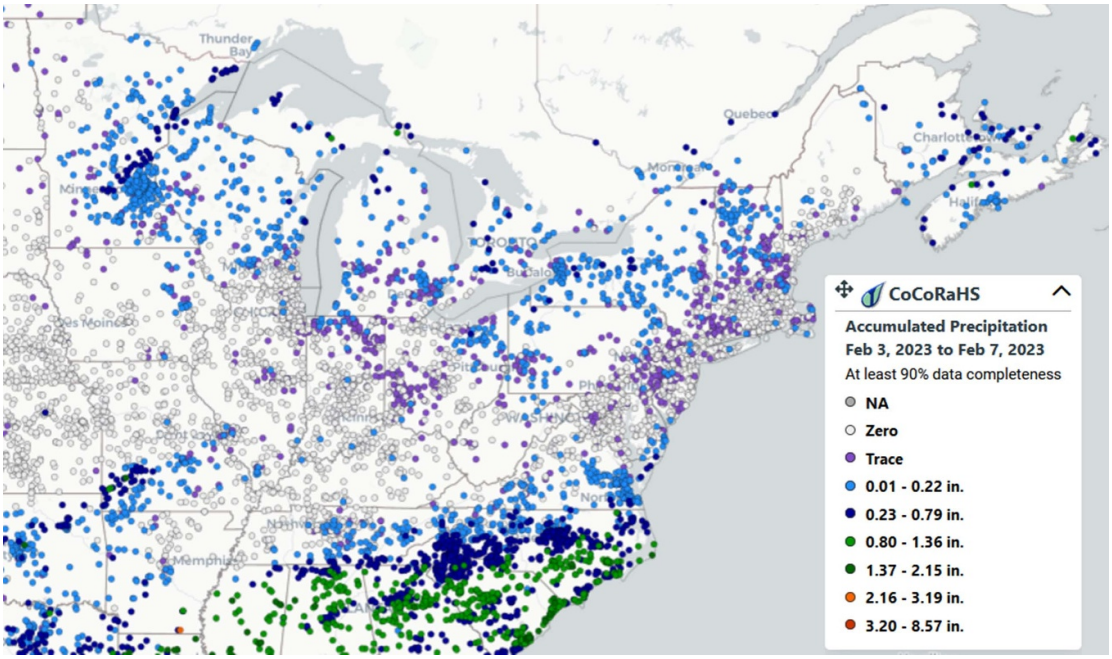

Figure 1. The location of the East Palestine train accident (star), and precipitation (inches) for 3 February (∼7 a.m.) through 7 February 2023 (∼7 a.m.) for the US Midwest and Northeast [10]. Reproduced from [10]. CC BY 3.0.

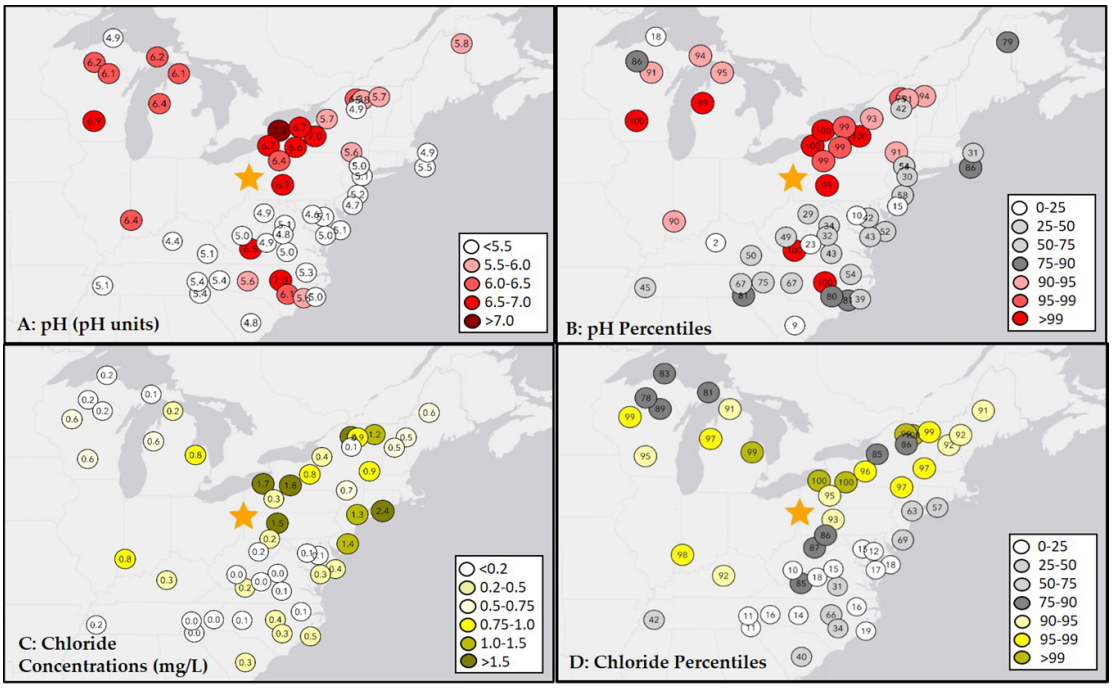

Figure 2. Wet deposition observations from 31 January through 7 February 2023 for (A) precipitation pH (pH units), (B) pH percentiles as compared to 2011 through 2021 wintertime observations, (C) chloride ion concentrations in precipitation (mg l^{-1}), and (D) chloride ion percentiles as compared to 2011 through 2021 wintertime observations. The location of the East Palestine train accident is marked with the star.

3.2. Chemical observations

In our review of the NADP-NTN analytical data, we expected to observe precipitation with lower pH and higher chloride values, given the likely emissions profile, and only find influences near the accident location (figure S1). We were surprised to find no unusually low (more acidic) pH values, but in fact found exceptionally higher (more basic) pH than normal (figure 2(A)). Typical annual average pH values for precipitation in the northeastern US are between 5.2 and 5.8 pH units [12], with winter pH values characteristically lower. For example, at site PA18 near the accident location, a typical winter pH is 4.9 pH units.

These anomalously high pH values were not restricted to the proximate accident location, but were widespread over the Northeast and Midwest. Figure 2(A) shows the accident week pH values measured by NADP, ranging from 6.4 to 7.3 pH units in the north and east, and near normal to the south. Values this high are rarely seen in the NADP weekly measurements, and never observed consistently over multiple sites in any one particular week. High values likely extended into southern Canada, but there are no NADP measurements here (see [13] for observations as available). Exceptionally high pH values also extended back into Michigan and Wisconsin, and similarly into Virginia and Indiana at a few locations. The interpretation of the two higher North Carolina values is complicated by high local emissions of ammonia and normally more basic pH values.

To put these unusually high values into perspective, we compared these accident week measurements of pH and all constituents to historic NTN values, and present the values as percentiles of the historic range (figure 2(B)). Percentiles were calculated for wintertime observations only (December, January, and February) for years 2011 through 2021 on a site-by-site basis. This accounts for any seasonality and long-term trends of the observations. A 100th percentile indicates the highest observation over this 11 year period.

From this analysis, it is clear that the pH values measured during the accident week were extremely high relative to historic data, and at 6 NTN sites actually represent the highest pH values measured over the period analyzed (figure 2(B)). Many more observations from the 14–16 state area were above the 90th percentile. This scale of observation alone at one or two sites may not be particularly compelling, but the sheer number of extreme percentages during this one-week period suggest a large impact to the atmosphere from the train accident over the Northeast and Midwest. The impact is especially clear over western Pennsylvania (near East Palestine), along the entire New York/Canada border, with decreased impact in northern Maine, and along the Eastern Seaboard (Nantucket Island did have unusually basic precipitation). Again, these consistently high pH values likely extended into southern Canada.

The same analysis was conducted for chloride ion concentrations (figures 2(C) and (D)). A similar pattern of exceptionally high chloride values is evident in the data. Values above the 90th historic percentile are frequent, with most from sites in the Northeast, but also Indiana, Michigan, and Wisconsin being historically very high. More normal values occur along the East Coast and into the southern states. From these two maps, it is very clear that extreme concentrations of multiple pollutants were present over a widespread area during the days after the accident, and resulted in enhanced deposition of these pollutants to aquatic and terrestrial ecosystems, including the Lakes Michigan, Erie, Ontario, and likely Huron and Superior.

This pattern of extreme concentration percentiles is also evident for the base cations, sodium, potassium, and calcium (Northeast in figure 3, and East in figures S3–S11). However, in-contrast, the percentiles for sulfate and nitrate were not as extreme as those of pH, chloride, and the base cations. The sulfate and nitrate values were high, but not as regionally consistent as the other compounds. For ammonium, very high concentrations were measured near the accident location and into New York. It is important to note that both sulfate and nitrate are secondary pollutants, or requiring atmospheric reactions from precursor gases, unlike the other pollutants.

Additionally, we observed that high percentiles for the same suite of chemicals (chloride, cations) extended into the week after the accident (7–14 February, not shown). However, the high percentiles moderated, and were generally further south into the Mid-Atlantic states. The upper Midwest concentrations return to a more normal range in the week after the accident, but many elevated pH values remained from Pennsylvania to Massachusetts, and well into Virginia. Sulfate, nitrate and ammonium levels were frequently high during the week after the accident, but not to the extreme extent as was measured with the other chemicals. See table S1 for all percentile ranks observed during the accident week.

From these observations, we can conclude that the major consequence from the accident was a widespread multi-pollutant impact over multiple states that extended beyond the accident week. We did not examine the third week after the accident in detail, but both the pH and chloride values were quite elevated at many sites, and particularly so in New York.

3.3. Meteorological observations and trajectories

Our precipitation chemistry observations are consistent with the state and flow of the atmosphere during and after the accident. First, the largest impact is seen at our NTN sites along the Canadian/New York border where precipitation (figures 1 and S2) occurred repeatedly from 3 February to 7 February. These sites were downwind of East Palestine during this period, and follow the predicted forward trajectories (figures 4 and S12(a)–(c)). Second, with the passage of the cold front on 5 February, air moved well into the southern US, to Virginia, Tennessee and Kentucky. The rain produced by the frontal passage likely then washed accident emissions out of the atmosphere at some of these sites. This flow into Virginia and North Carolina was confirmed with HYSPLIT forward trajectories (figures 4 and S12(a), (b)).

However, the impacts to precipitation chemistry in Michigan, Lake Michigan, and Wisconsin was

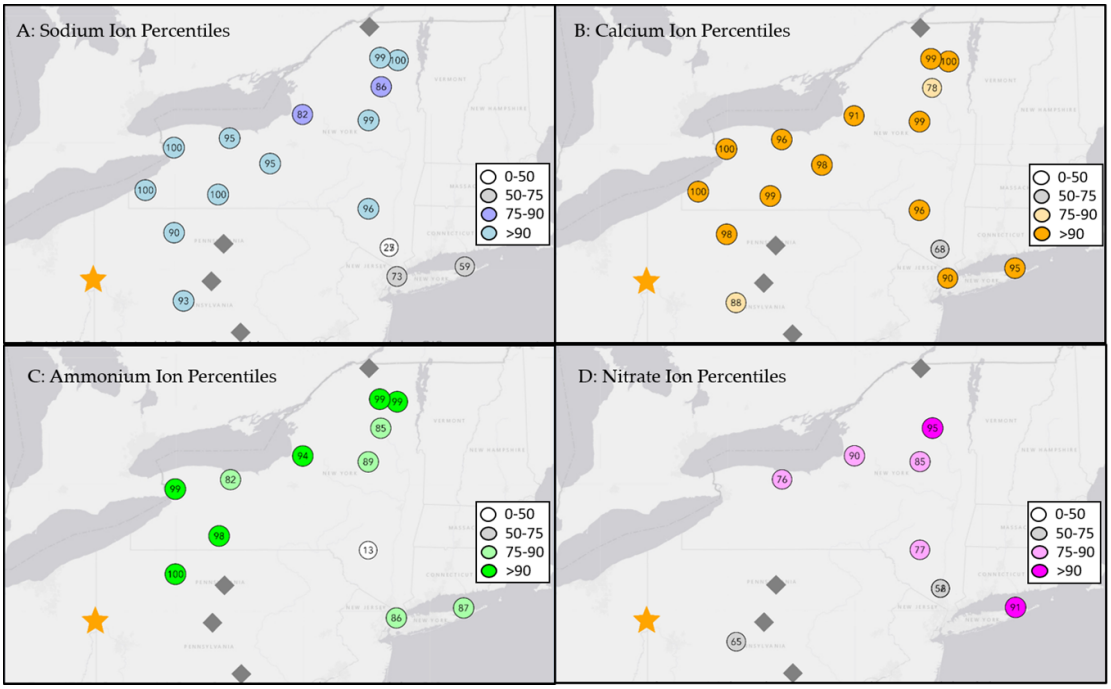

Figure 3. Wet deposition observations for New York and Pennsylvania from 31 January through 7 February 2023 for (A) sodium ion percentiles, (B) calcium ion percentiles, (C) ammonium ion percentiles, and (D) nitrate ion percentiles. Gray diamonds are NADP sites with no precipitation, and therefore no observations. The location of the East Palestine train accident is marked with the star.

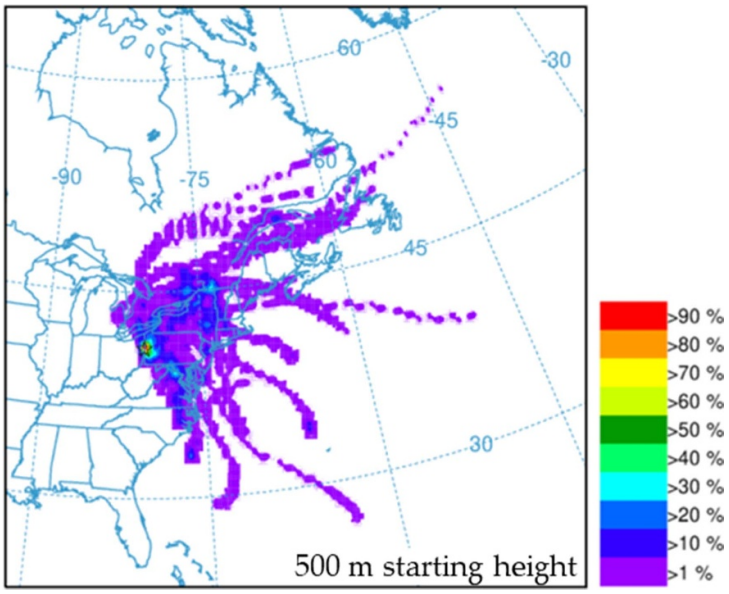

Figure 4. HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model forward trajectories for the accident week full fire period (1800 UTC 3 February 2024 through 1500 UTC 8 February 2024) for a 500 meter starting height and 48 h trajectory time. Color codes are the frequency of endpoints grid square/total number of trajectories (HYSPLIT output 'freq1', in %).

something of a surprise. Further detailed investigation of the meteorology showed air just above the surface moving towards the north-northwest at 6 p.m. on 6 February, as the controlled burn was concluding. A quickly moving low-pressure system was centered over Lake Michigan, associated with an occluding frontal system on that morning. Flow to the north/northwest and cyclonic flow (counter clockwise) at 925 millibars continued through to 6 a.m. on 7 February (figure S13), and likely moved pollutants into the upper Midwest region.

HYSPLIT forward trajectories showed air from the accident site clearly flowing north into Canada, and regional movement from central Ohio into

southeastern Michigan, but not into western Michigan and Wisconsin. It is possible that the HYSPLIT meteorological information did not capture the westward motion driven by the cyclonic flow present over Lake Michigan. Back trajectories from several points in western Michigan and southern Wisconsin during this late week precipitation event do show air originating in eastern Ohio, particularly at low levels (300 and 500 m, S12(c)), which does support that these extreme concentrations likely resulted from the accident. Further evaluation of surface observations for several locations in Michigan and Wisconsin on 6/7 February (Detroit, Saginaw, Grand Rapids, Milwaukee, Oshkosh) all show distinct surface flow to the west for consecutive hours between 6 February 10 a.m. through the morning of 7 February with high wind speeds (>15 miles per hour), followed by a distinct wind shift to eastward flow. All of this is quite consistent with a frontal passage, cyclonic flow, and widespread precipitation as the front passed (figures 1 and S13) [11, 14]. These observations argue that the conditions were in-place for accident pollution to move across Michigan and Wisconsin, and that precipitation removed the pollution as wet deposition as measured. Additionally, the Wisconsin and southern Michigan sites *only* experienced precipitation during the afternoon of 6 February, when cyclonic flow and contaminated air should have been present.

3.4. Likely sources of high cation concentrations and pH

High atmospheric chloride concentrations near the accident were expected, but the extremely elevated concentrations/percentiles of the base cations and high pH measured were surprising. Several studies have examined direct combustion of VCM under controlled conditions, and show overwhelming direct emission of HCl (ratio: 675 HCl:1 Phosgene at 950 °C) [15] and over 99% emission of Cl directly as HCl, but noted that more organic Cl forms could be emitted at lower temperatures (330 °C) [16]. It is important to note that anything but controlled conditions were prevalent during this accident, along with other non-VCM fuels simultaneously combusting. Therefore, oxidation of VCM after emission and emission of other organic chloride compounds (with subsequent atmospheric transformation) cannot be ruled out. Details about the accident relevant to chemical sources is only slowly becoming available, but we can make some initial interpretations of our observations from available information on fire response activities and the combusted cargo.

First, the train cargo burned during the accident included one single carload each of seminola (hard, Durham wheat), medical cotton balls, and frozen vegetables, among other products (table 1). As reported, these cars were full and completely combusted and we estimate total emissions of between 600 and 1100 kg of calcium, potassium, magnesium and sodium, using estimated cargo volumes, densities, and average cation concentrations [17–20]. Although these are rough estimates, it does show an important large and concentrated cation source (figure 3).

Second, between 22 and 26 February, news reports noted that PFAS-containing aqueous film-forming foams (AFFFs) were applied in the accident response, and that very large quantities of water used or impacted in the fire response is currently being disposed of through deep well injection due to elevated PFAS concentrations [21–24]. The National Transportation Safety Board hearings in June 2023 and related documents confirmed AFFF application in at least low amounts [25]. According to product specifications, this AFFF includes both PFAS and sodium sulfate compounds. Current AFFF formulations are also based in part on the addition of diutan gum/base cation mixtures [26]. If responding fire fighters used some type of common non-PFAS suppression dry powders or foams (Class A or B), which are usually based on sodium and potassium bicarbonate salts, this would also partially explain very high Na additions (observed) and high pH values (also observed), among other high cation concentrations. It is reasonable to conclude that the high cation precipitation concentrations were, in part, a result of injection into the atmosphere of fire suppressant cations applied over the multi-day accident response.

And *third*, high base-cation concentrations in precipitation are typically associated with elevated cation (calcium, sodium) carbonate (or oxides) compound emissions which result in the neutralization of acidity and thus higher than typical precipitation pH [27, 28]. Also, a very large volume of glycol compounds were completely combusted (5 train carloads or more), likely leading to substantial emissions of hydroxide ion (OH^-) in the fire plume (table 1), with further impacts on atmosphere/precipitation pH.

Clearly, the combustion of the train cargo and fire suppression employed provide many probable sources of the high cation concentrations and high pH observations seen downwind in the weeks after the accident. These explanations do not include other unknown emissions from the train cars and their contents.

3.5. Other explanations for high concentrations?

In wet deposition science, a well-known relationship is that low precipitation volume samples are associated with high ion concentrations, since the atmospheric pollutants are being removed with less precipitation. Lower concentrations are commonly

seen with high precipitation volumes, as noted by Lindberg, among others [29]. The generally low precipitation volume in the Northeast during the accident week contributed only marginally to higher chemical constituent concentrations as explained below.

First is the overwhelming presence of high pH (*lower* H^+ concentrations). In general, as precipitation volume decreases, lower pH (*high* H^+ concentration) would be expected. Therefore, the occurrence of so many high pH values (*lower* H^+ concentrations) argues against lower precipitation volumes leading to these high concentrations.

Second, high analyte concentrations were present regardless of the precipitation amount. Figure S14 shows all of the compound percentiles and precipitation amounts (volumes) for all New York sites. Site NY10, one of the closest to the accident, is one of the most extreme in percentile concentrations, but has the highest precipitation volume of all sites. NY99 (southeast New York) is also instructive; this site had very high precipitation volume (roughly equivalent to NY10), but had normal concentration percentiles and is likely south of the main accident emissions plume. NY93 (northern New York) also has extreme concentrations but also experienced higher precipitation. Other good examples are VA13, where almost 2 inches of precipitation occurred and has exceptionally high concentration percentiles.

Third, it is possible that the high Cl and Na came from road salt contributions as applied during the accident week over a broadly distributed area with snow (figure S2). We reviewed the historic (both winter and warm season) and accident week observations for a 1:1 Cl:Na ratio that would suggest major road salt contributions. Over all 11 historic winters, where road salt application is frequent, widespread and quite heavy, chloride and sodium levels never approached the extreme levels observed during the accident week (figure S15). Additionally, the Cl:Na ratio is typically between 1.1 and 1.2, is unlikely to be measured at 1:1, and can also go below 1. This suggests a much more complicated relation between Na and Cl. Some accident week ratios did move closer to a 1:1 ratio, but some ratios were higher and some ratios were less than 1:1. There was no consistent 1:1 ratio as would be expected if there was a large road salt contribution. Even sites with no/little snow and no salt addition in the area moved lower than normal conditions. Additionally, the accident week was generally a lower snow week over the multi-state area which argues that lower salting activities occurred. But, all accident week observations did show extremely large additions of both Cl and Na; this was consistent. Therefore, we must conclude that it is unlikely that these observations are caused by deicing salt, and that a high source emission of both Cl (from the VCM combustion) and Na ion occurred during the accident week.

A fourth possible explanation for these high concentrations would be that of widespread injection of aerosols from wildfire or forest fires. However, these types of fires are not common during the wintertime. A check for these conditions during early February showed that there were no major forest fires burning in the US, and that February 2023 was in fact lower than normal in the total number of fires and acres burned (50 000 acres) [30, 31]. This was also the case in Canada, where there were no major fires during February [32].

It is also conceivable that during the accident week, the 'normal' suite of sources over this area could have caused these observations: i.e. industrial activities and combustion, agricultural area sources, vehicles, etc. Undoubtedly, these sources contributed a fraction of the pollution burden for these normally observed chemical constituents. However, the extreme nature of these concentrations, seen at so many sites during this one week, argues that something besides the normal sources caused these historic winter concentrations, and that there must have been a separate multi-compound addition to the historic measurements. This extreme nature is clearly shown in figure S15, where this week stands above all other weeks in the 11 year historic period.

3.6. Overall summary observations

Integrating the observations of all compound concentrations and percentiles during the accident week, we sum the number of compound percentiles that were equal to or exceeded the 99th percentile to provide an overall estimate of the impact area. Figure 5 shows the sum of compounds at each site at or above the 99th percentile, and therefore the estimated areal extent of the accident (see figures S16, S17 for 90% and 95%, table S1 for all percentile ranks). From this, we estimate that the accident influenced a substantial portion of the US Midwest and Northeast, and extending into several southern states. In total, the impacted region encompassed parts of 1.4 million square kilometers or 14% of the US land area. At this level of certainty, 19 sites had at least one chemical compound in the 99th percentile, while 8 sites had 4 or more compounds in the 99th percentile.

In total, we estimate at least some impact on portions of 16 states and one third of the population of the US (110 million), and likely impacts to southern Ontario, and many of the Laurentian Great Lakes.

As a final check of reasonableness, we made a fairly simple estimate of the total excess Cl deposition to our study area resulting from the train accident. For this, we took all 20 sites with Cl concentrations above the 90th percentile during the accident week, and

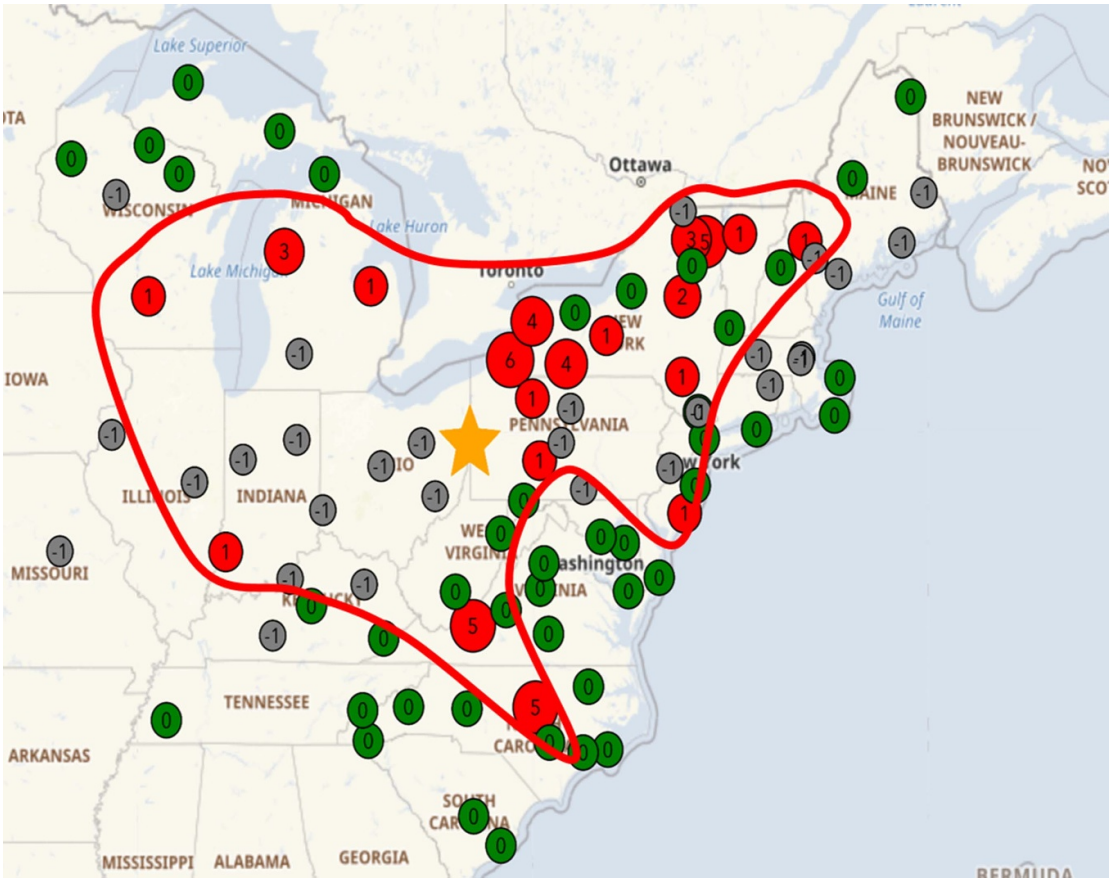

Figure 5. Summary area of impact, for the first week of the accident, based upon the sum of chemical analytes above the 99th percentile of historic measurement. Red indicators show the number of precipitation analyte concentrations above the 99th percentile. Sites with samples (green, '0') and without samples (grey, dry samples or '−1') are included.

subtracted away the 2021/2022 winter precipitation-weighted average site concentration. We then multiplied this value by the accident week site-specific precipitation amount to estimate excess chloride deposition at each site (mg m^{-2} $week^{-1}$). We then assumed a 10 000 sq. mile impact area to each site—which gave a reasonable approximation of total area, by state that had precipitation during the accident week. With this large number of simplifying assumptions, we estimated excess Cl deposition over the area equivalent to about 140% of the estimated mass of VCM combusted (see table S2 for specifics). Although marginally higher than the reported VCM mass in the tank cars, it is certainly within an order of magnitude of what was measured across the NADP network. It is certainly possible that there were other, non-VCM related Cl emissions from the fires. However, we do believe that this simplified mass-balance estimate bolsters our contention that the train accident fires contributed this chemical signal to a large surrounding area.

This is only an initial assessment of the impacts of the train derailment on atmospheric and precipitation chemistry, using publicly available NADP-NTN data; much more can be added to this understanding of the accident's impact. Additional study should focus on (1) a comprehensive modeling of the atmospheric dynamics and chemistry to more specifically show what pollutants were transported away from the accident location, where these concentrations were found, and how long these concentrations were present over a significant section of the US and Canada; (2) determine emission rates of the expected pollutants over time during the 5 accident days for further modeling; (3) examination of the atmospheric concentration measurements made by the Clean Air Status and Trends Network which measures rural concentrations of many of the same compounds in atmospheric aerosols; (4) a week-by-week investigation of the depositional differences of the pollutants to show overall flux increases; and (5) a full factor analysis to determine how uniform the high concentrations were over the affected area to provide additional support for the train accidents impact.

This paper does show the great value of a nationwide network for routine precipitation monitoring. Our observations allowed us to determine, initially, the regional atmospheric impact from the accident and subsequent response activities.

The NADP networks do not currently quantify specific organic compounds that might be more specific tracers of the train cargo, but given the widespread impacts to precipitation documented with currently measured NADP analytes, it is possible that there was a significant toxic organic flux to the earth's surface.

Data and materials availability

All measured data will be made available at the NADP website (https://nadp.slh.wisc.edu/networks/national-trends-network/) upon completion of the annual quality assurance and quality control of the network, following our normal operational procedures.

Acknowledgments

The NADP is the National Research Support Project-3: A Long-Term Monitoring Program in Support of Research on the Effects of Atmospheric Chemical Deposition. More than 250 sponsors support the NADP, including private companies and other non-governmental organizations, universities, local and state government agencies, State Agricultural Experiment Stations, national laboratories, Native American organizations, Canadian government agencies, the US Geological Survey, the US Environmental Protection Agency, the National Park Service, the National Oceanic and Atmospheric Administration, the US Fish & Wildlife Service, the Bureau of Land Management, the US Department of Agriculture—Forest Service, and the US Department of Agriculture—National Institute of Food and Agriculture under Agreement No. 2022-39133-38451. Any opinions, findings, and conclusions or recommendations expressed in this publication are those of the authors and do not necessarily reflect the views of the program sponsors or the University of Wisconsin-Madison.

Conflict of interest

The authors declare that they have no conflict of interests. *This work was partially funded by the* US Department of Agriculture—National Institute of Food and Agriculture under Agreement No. 2022-39133-38451, and the Wisconsin State Laboratory of Hygiene, General State Revenues.

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