# Attachment 18

T. David Hoyle, Esq, Member
Motley Rice, LLC
28 Bridgeside Blvd
Mount Pleasant, SC 29464

Ref: Expert Report Regarding Air Emissions from New Indy Catawba, LLC Mill

Dear Mr. David Hoyle,

This report summarizes my analysis of past and present efforts to monitor and report air emissions associated with the operations of the New Indy Catawba ("New Indy") pulp and paper mill located at 5300 Cureton Ferry Road, Catawba, York County, South Carolina ("Catawba Mill"), In my professional opinion, based upon the known, likely and potential emissions associated with pulp and paper mills like the Catawba Mill, these efforts have been and continue to be grossly inadequate and inadequately protective of the public health.

Emission monitoring to-date reveals that individuals and families residing around this mill have been exposed to levels of hydrogen sulfide well beyond those known to result in health effects. Current efforts to monitor and report hydrogen sulfide emissions, however, have failed and continue to fail to accurately capture the full extent of continued hydrogen sulfide emissions from the Catawba Mill, based upon limitations in the placement of air monitors.

Additionally, and critically, there has been no known investigation or monitoring whatsoever of other likely and/or potential emissions associated with pulp and paper mills like the Catawba Mill. The failure to monitor for these potential emissions presents severe health risks to nearby residents. In particular, there has been no monitoring or modeling of other potentially harmful TRS compounds, including methyl mercaptan, dimethyl sulfide, dimethyl disulfide, sulfur dioxide and other volatile compounds containing reduced sulfur, which are believed to constitute 90% of the TRS emissions at Catawba Mill. Additionally, there are numerous other air emissions associated with pulp and paper mills known to impact air quality and cause adverse health effects which have not been monitored at the Catawba Mill including: particulate matter (PM), nitrogen oxides (NOx), sulfur oxides (SOx), lead, carbon monoxide (CO), ammonia, *arsenic, *cadmium, chlorine, chlorine dioxide, cumene, *dioxins**,** *furans,

*formaldehyde, *hexavalent chromium, isopropyl alcohol, manganese, mercury, methanol, ethanol, methyl ethyl ketone, *polycyclic aromatic hydrocarbons (PAHs), phosphorus, selenium, sulfuric acid, and zinc. (Environment and Climate Change Canada, 2018) (Dionne & Walker, 2021). In addition, certain terpenes (i.e., resin acids (diterpenes) and vapors containing terpenes, including from turpentine) produced during paper manufacturing have been shown to pose a risk to skin irritation, allergy, and respiratory symptoms including asthma (Straumfors et al., 2018) As discussed herein, many of these emissions are known cancer causing agents, and have been associated with significant health effects.

**Known cancer causing agents (EPA, 2018).*

This report addresses multiple concerns about the lack of proper air quality impact assessment based on my review of the various documents, expert reports, presentations, peer-review literature, and other sources of information related to this case. (Appendix A). The statements and findings outlined herein are based on my education and experience as an environmental expert in air quality and human health exposure and risk assessment, including how environmental toxins from exposures result in both acute and chronic health impacts. The findings stated herein also draw upon my direct experience in designing and implementing several air quality and human health evaluations to assess downwind impacts of various air emissions. This prior experience in evaluating downwind emissions impacts includes determining an appropriate number of monitoring stations needed to yield reliable data, identifying scientifically reliable equipment in order to properly monitor the air emissions, and evaluating and assessing actual analytical results, including review of results for assessment of acute and chronic health outcomes in children and adults.

My opinions, analyses, and conclusions are provided with a reasonable degree of scientific certainty.

Biographical Sketch

I am the Founder & Chief Executive Officer of CHANGE Environmental, LLC a certified Veteran Owned Small Business engaged in Environmental Compliance and consulting services

headquartered in Saratoga Springs, NY. I am a recognized expert in the fields of air quality, epidemiology, infectious disease, and human health exposure and risk assessment. I have directed, managed, and consulted on numerous domestic and global environmental consulting and research studies dealing with exposures to various toxins, including gases, particles, viruses, mold. I have served as an expert witness on a variety of exposure cases, many of which included carrying out modeling, monitoring, and statistical data evaluations on populations for acute and chronic exposures.

1. *New Indy*

The ("Catawba Mill") is a pulp and paper facility commonly referred to as a Kraft Mill. By definition, a Kraft Mill refers to the "kraft process" (also known as kraft pulping or sulfate process) where conversion of wood into wood pulp takes place, which consists of almost pure cellulose fibers, the main component of paper. The Catawba Mill manufacturers brown paper also referred to as linerboard. As defined in other reports reviewed, this process generates significant malodorous and toxic emissions (including hydrogen sulfide and other TRS and volatile organic compounds). If not properly controlled through the use of recognized methods (e.g., steam strippers), emissions of these compounds can be very strong and have a significant impact on local and regional air quality both for acute and chronic exposures.

1. *Air Quality Assessment at New Catawba, LLC Facility*

As noted, prior expert reports, presentations, and documents were reviewed related to the assessment of emissions characterization of hydrogen sulfide and lack of information for TRS compounds with my expert report adding additional information on the various other environmental toxins that impact air quality surrounding pulp and paper mills, that includes the Catawba Mill. This also includes a detailed review of the October 2021 Air Dispersion Modeling report carried out by the Catawba Mill. This report will not re-evaluate the findings of the various expert reports (Meggs, Norcross, Hanna, and Osa) [2] but is in agreement with these expert assessments and their conclusions regarding the lack of proper air quality modeling and full impact assessment on local and regional air quality. I have outlined below the other

significant deficiencies and data gaps remaining which, if left unaddressed, will vastly underestimate the actual impact of the emissions from the Catawba Mill on the local communities. This includes the understanding and knowledge that modeled results and back calculations indicate that for several months in 2021 emissions from the Catawba Mill were likely 1,500 times greater than reported. With the failure of New Indy and EPA to require the monitoring and modeling of TRS and other known environmental toxins outlined above, the true full extent of exposure and health outcomes remains largely underestimated.

*2. Air Quality Monitoring at The Catawba Mill*

The air monitoring program implemented here, based directly on EPA input, suffers from data gaps and inadequate logistics, and fails to yield proper air quality impact assessments related to the Catawba Mill. These deficiencies render this monitoring program inadequately protective of the public health, as this monitoring program (1) fails to accurately report hydrogen sulfide emissions, based upon the number of monitoring sites and their locations; and (2) fails entirely to monitor and report other likely and/or potential air emissions, many of which are known to result in health effects, including other TRS constituents and other harmful emissions.

A detailed review of the October 2021 air dispersion modeling report regarding the Catawba Mill was conducted. Various reports have clearly identified issues with that report and therefore, will not be re-evaluated here. I agree with the comments presented (e.g., Hanna, Meggs, Norcross, and Osa reports)[3]. As previously noted, the fact that emissions of neither TRS compounds nor the several additional environmental toxins listed above were ever evaluated creates a significant data gap and are contributing factors to widely reported smell and human health effects.

*[3]Richard Osa, September 24, 2021, Dr. William Meggs, September 26, 2021, Richard Osa, December 23, 2021, Dr. Stephan Hanna, December 2021, Kenneth Norcross, September 24, 2021 and December 2021*

To properly identify and quantify if the placement of both the fence line and community monitoring stations were rational, it became important to understand a more detailed multi-year meteorological assessment of wind speed and direction in the region as air pollution migration, deposition, and dispersion are highly dependent on local wind speed and direction. Reports reviewed do provide very discrete information on individual days and smaller breakdowns of local wind speed and direction, and although important for specific events and assessments, they are not able to show all variations of wind speed and direction. For example, Dr. Hanna's report indicates that on April 27, 2021 "winds were fairly steady out of the SSW." On that day, this correlates with the findings that the dominant wind direction is from the S-SW to the N-NE direction, which for his assessment was important and informative. However, the wind rose map clearly indicates that there are also varying directions to where winds blow to. Based on this assessment and lack of any previous analysis by New Indy or EPA, the placement of monitors, both at the fence line and in the surrounding community, raised very serious concerns, namely, that air emissions of hydrogen sulfide, other TRS components & various other environmental toxins (not monitored at all) would likely not have been captured and representative. This was a major issue that was not highlighted nor described in the Quality Assurance Project Plan (QAPP). Previous reports [3](e.g., Osa, September 2021) do correctly advise that given the size of the property, the number of fence line monitors (3) would not be sufficient, nor accurate placement of the monitors as shown in Figure 2.

*[3]Richard Osa, September 24, 2021, Richard Osa, December 23, 2021*

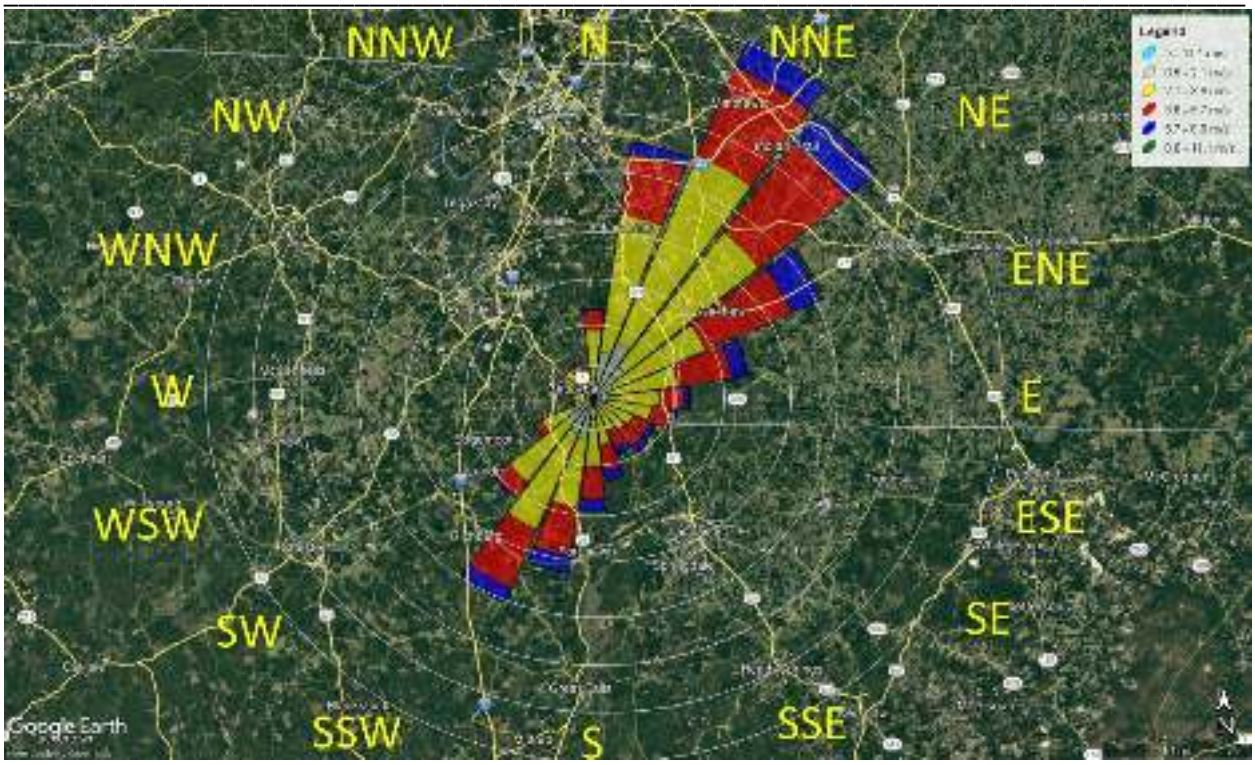


Figure 1. Wind flow vectors plot (blowing to).


Figure 2. Catawba Fence Line Monitoring locations as designated by EPA.

In assessment of air emissions and exposures it is important to remember that plume migration, dispersion and deposition as stated above is driven heavily by local wind speeds and direction. Air pollutants not only move horizontally upon emission, but there is also a vertical dispersion and migration of emissions which, without sufficient characterization and placement of monitors, may, if not accounted for in the setup of monitoring and modeling protocols, will result in vastly underestimated emissions. Comparison of the wind rose map (tool used to assess wind speed and direction at a particular location) to the placement of the three

-fence line monitors[4] does not justify placements at Site 1 or Site 2.

*[4]As detailed herein, in order to gather reliable data, several more monitors in varying directions on the property line should have been deployed.*

These sites as illustrated would not capture the fullest extent of emissions from the Catawba Mill or its wastewater treatment processes in an accurate manner, given a low percentage of the wind blowing in the direction of the monitoring stations, and predictably fail to obtain robust data on the levels of hydrogen sulfide (or other, unmonitored components/emissions) leaving the facility when compared to data drawn from nearby community monitors. Site 1 for example, is not only further from most of the community monitors but does not accurately capture representative emissions of the Catawba Mill's even accounting for varying winds from the N-NE to S-SW as it is allocated SE of the facility. Similarly, given that the monitoring station at Site 2 is located directly east of the Catawba Mill, based on the wind rose data it would not be in a location that would be representative for capturing a robust emissions profile from the Catawba Mill.

In addition, it is not clear why there is no upwind monitoring sites, both at the fence line and in the community. These are critical to provide comparative analysis should there be any major shifts in wind direction and speed directly impacting typical downwind emissions and constitutes a major hole in data used in air dispersion analysis and interpretation of boundary conditions in emissions inventories. Materials reviewed to date fail to identify any justification

from New Indy or the EPA for employing a monitoring technology that, by design, provides an output with material gaps in the emissions data collected.

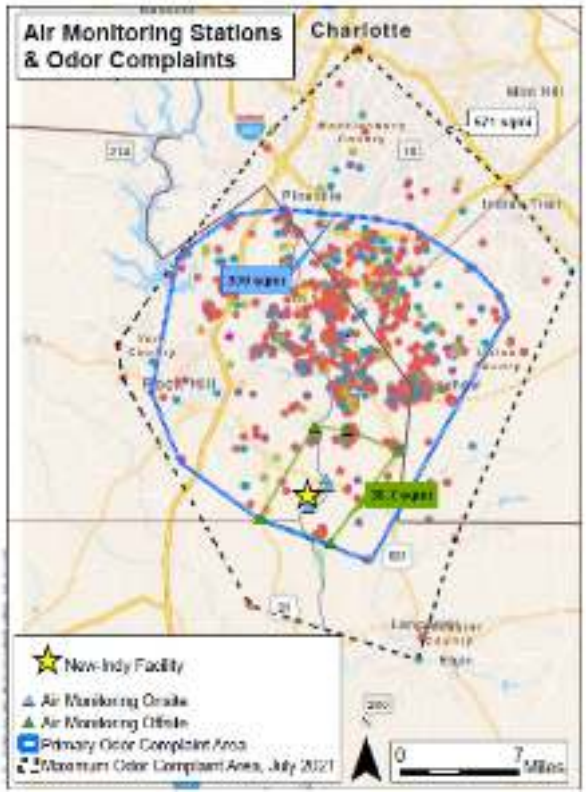


Figure 3. Air Monitoring Stations and Odor Complaints

As part of my detailed analysis, I reviewed the areas with odor complaints against the location of the monitoring stations as well as the wind direction assessment (figure 1). This analysis highlights strong concerns for the rationale used for placement of the monitoring stations by New Indy and EPA when considering the vast spatial variability of the complaints presented. As indicated earlier, plume migration resulting from vertical and horizontal dispersion can have a strong temporal variability even under small shifts in wind direction and speeds yielding anywhere from short term to long term exposures. It is critical to perform an assessment to understand dominant downwind wind directions, but to also take into consideration the characterization of smaller shifts in all surrounding areas, as winds do not only blow statically in

the dominant directions. This is not the case as indicated by the placement of the monitors shown in figure 3. Reports clearly show complaints in all directions of the facility and at varying distances, indicating plume migration and dispersion across multiple directions.

In review of the EPA community monitoring network there are neither fixed sites west of the facility nor S-SW of the facility to capture emissions when winds are blowing in that direction, despite complaints in all directions of the facility. This leaves large issues of uncertainty that would not exist if all areas of exposure were being properly monitored.

Overall, despite being provided maps of fence line and community monitoring programs, the locations of the sites, including the lack of adequate fence line monitoring (use of only 3 sites), as approved by New Indy and EPA, draws significant concerns as noted above. On a more general note, it is unclear what efforts were made to calibrate and/or assess the reliability of these measurements to ensure data credibility and quality assurance. A June 8, 2021, memo from Pete Cleveland, states, "New-Indy's consultant is checking and validating the daily precision checks for each station" including overall precision testing and simply relying on precision testing from 0-10% and if within this range would not require recalibration. It is not clear how this is being done and by whom. It is also unclear if EPA was also coordinating one of their own staff to provide third party comparative analysis and measurements to ensure all equipment at all the sites was operating in full functionality. This is even more critical especially with monitors running continuously as drift in readings can result in underreporting of actual ambient concentrations.

In addition to likely underreporting hydrogen sulfide emissions resulting from deficiencies in monitoring number and placement, the complete failure to monitor for any air emission other than hydrogen sulfide similarly defies logic. Hydrogen sulfide makes up only 10% of their total emission with TRS about 90% and no monitoring for other known environmental toxins outlined (Table 6.1, July 2021 New Indy Corrective Action Document). With EPA only requiring testing for hydrogen sulfide vastly undermines the integrity of the entire fence line, off site, and community monitoring programs by not capturing all emissions directly involved in operations

of Catawba Mill. In addition, the EPA sets standards that are supposed to be protective of human health and the environment. The work carried out under EPA direction at the Catawba Mill greatly undermines the integrity of the EPA's message to the public and its concern against acute and chronic exposures to very toxic compounds many of them not monitored, modeled, or even discussed.

*3. Air Pollution Health Effects & Exposures to Pulp and Paper Emissions*

As set forth above, due to failures in the current air monitoring program, available data concerning hydrogen sulfide emissions from the Catawba Mill is not accurately representative of the emissions likely impacting residents around the Catawba Mill. Further, the current air monitoring program fails to monitor for any other toxin known to be associated with emissions from pulp and paper mills like the Catawba Mill whatsoever, including other TRS constituents (e.g., methyl mercaptan, dimethyl disulfide, dimethyl sulfide and other sulfur oxides). Various other environmental toxins noted herein to which exposure can yield short and long term health effects supported by research studies that including particulate matter (PM), nitrogen oxides (NOx), sulfur oxides (SOx), lead, carbon monoxide (CO), ammonia, *arsenic, *cadmium, chlorine, chlorine dioxide, cumene, *dioxins**,** *furans, *formaldehyde, *hexavalent chromium, isopropyl alcohol, manganese, mercury, methanol, ethanol, methyl ethyl ketone, *polycyclic aromatic hydrocarbons (PAHs), phosphorus, selenium, sulfuric acid, certain terpenes and zinc. (Environment and Climate Change Canada, 2018). Many of these chemicals are cancer causing agents (EPA, 2018).

The failure to monitor for these other constituents is particularly problematic here, where residents around the Catawba Mill continue to report strong odors, at times distinct from the characteristic "rotten egg" smell of hydrogen sulfide. These reports indicate that these residents are likely being exposed to other as yet unmonitored and unknown air emissions arising from operations at the Catawba Mill, presenting a significant potential public health risk that must be mitigated through implementation of a proper air monitoring program.

a. The current monitoring program fails to monitor for any TRS constituent other than H2S, neglecting emissions which present greater toxicity to the public, and constitute a much larger share of New Indy's overall emissions.

Research has shown that exposures to TRS from pulp and paper operations causes adverse health effects, including difficulty breathing resulting in increased hospital visits. Kangas et al., assessed concentrations of hydrogen sulfide, methyl mercaptan, sulfur dioxide and its derivatives in kraft mills. Exposures ranging from 0-20 ppm hydrogen sulfide, 0-15 ppm methyl mercaptan and similar levels for dimethyl disulfide and dimethyl sulfide all resulted in increased health outcomes (e.g., headaches and decreased cognitive function). Impacts from TRS exposures have been found to be elevated in sensitive populations (e.g., asthmatics, children) (Campagna, 2004). The significant risk of health effects from TRS compounds other than hydrogen sulfide is reflected in much lower fence line limits for constituents like methyl mercaptan than for hydrogen sulfide as imposed by DHEC, (20 ppb) v. hydrogen sulfide (140 ppb), and similarly, in allowable workplace standards for TRS components like methyl mercaptan, which are half of allowable concentrations of hydrogen sulfide (10 ppm vs. 20ppm). (Occupational Safety and Health Administration (OSHA) Personal Exposure Limits for Methyl Mercaptan and Hydrogen Sulfide) . Despite, this, there had been no known investigation or monitoring of methyl mercaptan concentrations or other TRS constituents in the regions surrounding the Catawba facility. Not accounting for these emissions is a major flaw and data gap in their analysis and vastly under predicts overall impacts on health effects from emissions.

New Indy's air dispersion modeling report perpetuates these data gaps, failing to account for methyl mercaptan and other TRS compounds in the modeled analysis. This makes it impossible to accurately predict the full extent of health outcomes resulting from TRS impacts that are emitted from the Catawba Mill and its Wastewater Treatment Plant as detailed and described elsewhere.

b. The current air monitoring program fails to consider other known emissions associated with pulp and paper mills like the New Indy Catawba facility, which present their own significant potential health effects to residents.

In addition to TRS emissions, a variety of other air emissions are associated with pulp and paper operations like the Catawba Mill. These include particulate matter (PM), nitrogen oxides (NOx), sulfur oxides (SOx), lead, carbon monoxide (CO), ammonia, *arsenic, *cadmium, chlorine, chlorine dioxide, cumene, *dioxins**,** *furans, *formaldehyde, *hexavalent chromium, isopropyl alcohol, manganese, mercury, methanol, ethanol, methyl ethyl ketone, *polycyclic aromatic hydrocarbons (PAHs), phosphorus, selenium, sulfuric acid, certain terpenes and zinc. (Environment and Climate Change Canada, 2018) (Straumfors 2018). Where, as here, residents are subject to continuing but as yet unidentified emissions from the New Indy facility, further investigative monitoring and assessment of these potential emissions is critical given their potential, significant impacts on human heath.

**Known cancer causing agents (EPA, 2018)*

Many of these compounds have been identified as known, probable or possible carcinogens, with others having been identified as the cause of respiratory and cardiovascular symptoms. For example, (White et al and Birnbaum 2009) described the impacts of *dioxins and dioxin like compounds* and exposure at early age resulted in neurologic, immunologic, and reproductive issues and that all data from an epidemiological standpoint clearly outlines humans are not be subject to exposure to these cancer-causing agents. *Formaldehyde exposure* has been understood for decades to lead *to* increased rates of cancer development through inhalation (Swenberg et al., 2013). It is also known and documented that *Cadmium* poses risks that are not only associated with occupational exposures, but environmental exposures that have been shown to directly impact human health and genome (Hartwig, 2013). *Polyaromatic Hydrocarbons* have been shown to result in lung cancer and are responsible for the highest mortality in the United States with both occupational and environmental exposures resulting in high incidence of cancer development (Moorthy et al., 2015). Inhalation and ingestion from

food or water of *Hexavalent Chromium* is well documented to affect human health even direct contact from skin. Lung cancer from exposure via inhalation is well documented in adverse health outcomes and lung cancer (National Institute of Environmental Health Sciences, 15th Report on Carcinogens, 2021). Since the early 1990's a significant amount of research has been conducted around *particulate matter* exposures and its impact on respiratory and cardiovascular disease with primary focus on particulate matter of diameter <2.5 um given its ability to penetrate deep into the lungs resulting in human health effects (Prabhakaran et al., 2020).

Where, as here, residents report continuing odor complaints, it is critical to conduct further investigation to identify potential toxins that may be emanating from the New Indy facility, to ensure that they are not presenting unacceptable risks to human health – particularly where many of these toxins have lower concentration threshold for enhanced health effects than hydrogen sulfide.

c. Current H2S requirements fail to protect the public health, and representations from the expert retained by New Indy fails to accurately describe the potential risks associated with these exposures.

The Agency for Toxic Substances & Disease Registry (ATSDR) designates hydrogen sulfide Minimum Risk Level (MRL) for Acute Inhalation as 70 parts per billion (ppb) for exposure from 1-15 days, intermediate MRL of 20 ppb for exposures from 1-365 days, and chronic exposure reference dose of 1 ppb for lifetime. The EPA required Catawba Mill to comply with the 70 ppb MRL benchmark based on rolling 24-hr 7 day averaging period and 600 ppb in any 30-minute period. (May 2021, EPA Clean Air Act Emergency Order).

These limits would not correlate to the protection of public health especially for sensitive populations. This is supported by the study noted above by Campagna, 2004 that found increased hospital visits and asthma exacerbation if hydrogen sulfide and TRS levels exceeded 30 ppb for any 30-minute period. This is 20 times lower than the 600 ppb 30-minute limit and half of the 70 ppb rolling average set by New Indy and EPA for protection of public health. In

addition, the World Health Organization based on data on respiratory issues and odor annoyance recommends hydrogen sulfide levels not exceed 5 ppb during a 30 min period which is 120 times lower than the required 600 ppb level and nearly 15 times lower than the 70 ppb rolling average at Catawba Mill. Additional research (Marttila et al., 1995) has shown that occurrence of exposures to TRS ranging from 0 to 58 ppb, was associated with ocular, respiratory, and neuropsychological symptoms including increased nasal and pharyngeal irritation in communities near Pulp and Paper Mills.

A May 2016 publication by Finnbjornsdottir et al., at Center of Public Health Sciences, University of Iceland, "Association between Daily Hydrogen Sulfide Exposure and Incidence of Emergency Hospital Visits: A Population Based Study" demonstrates that the MRLs provided above are not sufficient for protection of public health with respect to low level modeled ambient concentrations of hydrogen sulfide. The source of hydrogen sulfide resulted from a nearby geothermal power plant. Short-term associations of exposure higher than 5 ppb yielded a statistically significant outcome of exposure and emergency room visits based on unstratified models with a mean age of 70 years (e.g., cardiovascular & respiratory distress). Additional research noted by Dr. Meggs showed that concentration of hydrogen sulfide greater than 30 ppb yielded increased hospital admissions related to asthma incidence in children.

The results of this study clearly indicate that acute low-level exposures to hydrogen sulfide higher than 5 ppb were dramatically responsible for reported health outcomes that again occur at levels 120 times lower than the approved 30-minute rolling average and almost 15 times lower than the 70 ppb 24-hr rolling average. The level of exposure and health is nearly 15 times lower than the acute MRL set forth at 70 ppb for rolling averages, 120 times lower than the 600-ppb level associated with 30-minute exposures, and 4 times lower than the 20-ppb intermediate MRL of hydrogen sulfide exposure. From these results and in comparison, the supposedly protective MRL set for the Catawba Mill's hydrogen sulfide levels are not to be considered safe nor at all protective of public health for reasons noted above.

This is also supported by the data set provided by Monitoring Station 1 (south of the Catawba Mill). In 2021 from May 26-31 and June 7-17 hydrogen sulfide levels were all above the 70 ppb 24 hour rolling average, ranging from 70.8 ppb to 207 ppb. On June 4, 12, 14, 15, and 20$^{th}$ all readings for hydrogen sulfide were higher than 600 ppb 30-minute rolling average, ranging from 607 ppb to 1330 ppb. These data clearly show that even with the unprotective limits set by EPA for the Catawba Mill, there were significant exceedances. If more stringent and protective limits were put in place, the levels of exceedance at the monitoring stations would have been much higher.

I have reviewed June 3, 2021 letter by Dr. Christopher Teaf that provides a 5 day wind rose plot. It is not clear what is being defined in his analysis as he mentions nothing in his report supporting the wind rose analysis and its relationship to defining the fence line monitoring program. From the review he states, "Once a more robust database is available, it would be appropriate to use the 140-ppb adjusted MRL value for decisions regarding property line hydrogen sulfide concentrations simply based on ceiling limits set by AIHA ERPG-1 reference and OSHA." This directly contradicts several research findings discussing levels of hydrogen sulfide yielding acute and chronic health outcomes as discussed above. Implementing a higher limit of 140 ppb would only result in expanded exposures and higher limits for release of hydrogen sulfide further impacting the community. Without a more robust and fully comprehensive data assessment, it would be highly premature and not justified to increase the fence line allowable concentrations given already reported health outcomes at much lower concentrations. Based on available data presented in various reports and emissions impacts across the region, a stricter fence line concentration would be warranted for hydrogen sulfide. More importantly, Dr. Teaf does not at all recognize in his analysis that the larger percentage (90%) of the total TRS emissions are TRS compounds (e.g., methyl mercaptan, dimethyl disulfide, dimethyl sulfide and other sulfur oxides) other than hydrogen sulfide.

I have also reviewed a recent questions and answer session (Research, Inc Interview) by Dr. Teaf and found some of his inputs to questions a bit alarming and not accurate. For example, he states, "The fact that you are smelling an odor does not mean that it is causing you harm." This

statement is false and does not adjust for desensitized members of the population that may have conditions that affect their olfactory sensory detection for hydrogen sulfide and TRS especially at very low odor thresholds from 2 ppb (ATSDR) and 5 ppb (WHO) that coincide with similar findings of levels lower than recommended by New Indy and EPA for protection of public health. A summary of health effects across the various environmental toxins via direct exposure to one or all together (symptoms within sensitive populations (children, elderly) can result in elevated impacts to exposures) include the following:

- Aging
- Burning/Itchy Eyes
- Exhaustion/Fatigue
- Nausea/Vomiting/Fever
- Upper and Lower Respiratory Distress, Asthma Symptoms and Exacerbation
- Systemic & Peripheral Inflammation
- Dermatitis
- Circulatory Complications
- Blood Disorders
- Immune system depression
- Development of Various Cancers
- Growths in the nasal cavities
- Head injury
- Hormonal disturbances
- Dental problems
- Lack of Coordination

- Tremors
- Seizures
- Myocardial Infarctions
- Multi-organ Impairment
- Pulmonary Edema

Therefore, just because you cannot smell it does not mean it cannot hurt you as detection cannot dictate awareness and therefore yield prolonged exposures that otherwise under normal circumstances may be avoidable.

Dr. Teaf also makes mention of the total deaths in the United States from H2S based on US Bureau of Labor Statistics, concluding that there have been "fewer than 50 fatalities" from 2011-2017. His analysis focusing solely on occupational settings. It is also important to consider that various organizations focusing on hydrogen sulfide exposures have shown having prolonged exposure to hydrogen sulfide gas can cause unconsciousness that can lead to headaches, reduced attention span and motor functions including other health issues. For example, heart issues can occur with exposures not yielding health effects until sometimes up to 72 hours following removal from the affected environment. In addition, delayed pulmonary edema, a buildup of excess fluid in the lungs, may also occur. Therefore, depending strictly on actual guidelines and determination of health outcomes is not appropriate in dealing with air toxins and acute and chronic effects. Most importantly these are just specific to hydrogen sulfide and do not address the combination of hydrogen sulfide that would be additive to the 90% TRS that would all together have significant short- and long-term impacts along with the extensive list of environmental toxins that are also known to be part of pulp and paper mill operations. This is also recognized by Dr. William Meggs in his September 2021 report, where he states, "*When there are complex mixtures with synergistic toxicities such as the hydrogen sulfide, methyl mercaptan, methyl disulfide, and dimethyl disulfide that plaintiffs were exposed to, there are no established minimum risk level that would be below quoted levels for individual*

*exposures."* In agreement with Dr. Meggs, air emissions related to Pulp and Paper Mill emissions are comprised of complex mixtures that together yield acute and chronic health impacts as noted herein.

One additional statement made by Dr. Teaf that warrants pause is: *"If you are outdoors when you smell hydrogen sulfide you may wish to go indoors temporarily to avoid the smell."* Major concerns with that statement are gaseous materials have penetration efficiencies of roughly 1, which indicates gases are able to penetrate the building envelope with little interference and impact the indoor environment even if windows and doors are all closed. This escalates with any open windows or doors. In other words, unlike particles where size and distribution play a role in penetrating building envelopes, gases act without much interference. Second, this also assumes that anyone going indoors to avoid the smell would have all windows and doors closed, which may not be case, and which could thereby increase the level of gaseous vapors from the hydrogen sulfide indoors. Third, assuming homes do not have any means of filtering the air, persistent exposures to hydrogen sulfide can exist in air from 15-45 days, especially with moderate temperatures and consistent source strength can continue to persist in exposures increasing health effects and outcomes over time.

Finally, I have also reviewed the PowerPoint presentation made by Drs. Abby Mutic and Melissa Gittinger on behalf of the EPA. As a preliminary matter, this presentation discusses only the health impacts of hydrogen sulfide, failing to fully account for the potential health risks posed by New Indy's various other emissions, including other TRS constituents, as described more fully above. Second, this presentation acknowledges that the health impacts of hydrogen sulfide are not fully understood at chronic levels. Permitting New Indy to continue to emit these harmful compounds creates known health risks, as well as currently unknown future potential health consequences. Under these circumstances, public health interests must be prioritized. Finally, the EPA presentation failed to address more recent studies showing health effects of hydrogen sulfide at low/chronic levels, as in *Campagna* et al. and Finnbjornsdottir et al.

*Closing Remarks*

Given my experience and expertise as an expert in air quality, human health exposure and risk assessment, New-Indy and EPA have failed to protect the environment and public health for those subject to the odors and adverse health effects resulting from the operations of the Catawba Mill. This is evident as supported by the various points made in this report that critically evaluate and provide disapproval for the actions of New Indy and EPA, especially considering that EPA has required the Catawba Mill to monitor and model only for hydrogen sulfide, even though it has been identified and documented as a minor input into the overall exposure paradigm at 10% of TRS impacts. The identified deficiencies in this air monitoring program includes not only improperly setting up monitoring and modeling for hydrogen sulfide, but failing to account for all the other known emissions related to pulp and paper operations as described. These data gaps substantially put the health and wellbeing of the residents in the community at risk for more chronic impacts to the poor air quality impacting the community. This is evident based on the video that I watched where various citizens are impacted by New Indy and are suffering from adverse health impacts to which match those symptoms described in this report and that are related to the operations of the Catawba mill.

All opinions provided in this report are made with a strong degree of scientific certainty based on experience and expertise.

Timothy R. McAuley

Name:

Timothy R. McAuley, MS, PhD

March 11, 2022

Date

Appendix A

List of Documents Reviewed

1. Richard Osa, September 24, 2021
2. Dr. William Meggs, September 26, 2021
3. Motion to Intervene and Lift Stay, September 29, 2021
4. Consent Decree, December 29, 2021
5. EPA Kraft Pulping Paper, March, 1979
6. Pete Cleveland, June 28, 2021
7. Dr. Christopher Teaf, June 3, 2021
8. Complaint in Intervention, September 29, 2021
9. Dr. Muttic & Dr. Gittenger Presentation
10. Dr. Christopher Teaf, Q&A Interview
11. Supplement in Support of Intervene and Lift Stay, December 23, 2021
12. Richard Osa, December 23, 2021
13. John Wilhelmi (ERG)
14. New Indy Catawba, Corrective Action Plan, Air Dispersion Analysis, October 2021
15. Dr. Stephan Hanna, December 2021
16. Kenneth Norcross, December 2021

List of Publications/Reference Materials Reviewed

1. Finnbjornsdottir et al., Association between Daily Hydrogen Sulfide Exposure and Incidence of Emergency Hospital Visits: A Population-Based Study. PLoSOne (2016)
2. Dionne and Walker. Air Pollution Impacts from a pulp and paper mill facility located in adjacent communities, Edmundston, New Brunswick, Canada and Madawaska, Maine, United States. *Environmental Challenges (2021).*
3. Environment and Climate Change Canada, Historical Substance Report (2018)
4. Campagna and Kathman et al., Ambient hydrogen sulfide, total reduced sulfur, and hospital visits for respiratory diseases in northeast Nebraska, 1998-2000. *Journal of Exposure Analysis and Environmental Epidemiology*. (2004)
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**Timothy R. McAuley, MS, PhD Curriculum Vitae**

Dr. Timothy R. McAuley is the Founder & Chief Executive Officer of CHANGE Environmental, LLC a certified Veteran Owned Small Business headquartered in Upstate, NY. Dr. McAuley is a leading authority and multi-award-winning environmental leader and recognized expert in the fields of air quality, epidemiology, infectious disease, and human health exposure and risk assessment. He has directed, managed, and consulted on numerous domestic and global environmental consulting and research studies in his areas of expertise. As a result of his work and contributions to the environmental field, Dr. McAuley has been invited to give several keynote speeches and has become a global resource for environmental forward thinking and a leader in his field. He is also currently an elected member by his peers to several national committees.

Dr. McAuley is a highly sought-after expert for given his extensive background and strong record in epidemiology and infectious disease along with being a leader in his field around air quality and human health exposure assessment. He has been supporting several clients (attorney's/private clients) recently and over the years on viral (e.g., COVID-19, SARS), bacterial, mold, and various impacts on health (i.e., inhalation, dermal, and ingestion exposures).

Dr. McAuley also provides additional consulting and expert services to dozens of. attorneys across the United States on several high profile dealing with various environmental impacts. Dr. McAuley experience and expertise stretches across 17 years for cases supporting attorneys for plaintiffs, defense teams, community groups, and non-profit organizations.

Dr. McAuley received his PhD. in Environmental Science and Engineering and his MS in Chemistry Clarkson University along with a BS in Biochemistry from The College of Saint Rose, where he is also a member of the Board of Associates and a member of the Alumni of Prominence Cabinet recognizing individuals who have demonstrated exceptional leadership and dedication to their industries.

EDUCATION

2006 PhD, Air Quality, Human Health Exposure and Risk Assessment , Clarkson University, Potsdam, NY

2003 MS, Chemistry, Clarkson University, Potsdam, NY

2001 BS, Biochemistry, The College of Saint Rose, Albany, NY

HONORS AND AWARDS:

2020 & 2021

- CHANGE Environmental, LLC Best National Environmental Strategy and Compliance Consultancy Award
- Albany Business Review Professional Achievement Award for Environmental Consulting and Leadership

2019

- National Renewable Energy and Policy Climate Change Award, by Environmental Business Journal International.

2017

- National Community Engagement and Business Achievement Award, by Environmental Business Journal International.
- Awarded, Inclusion into Industry Experts Magazine

2016

- Awarded, National Environmental Leadership and Excellence
- Award by Environmental Business Journal International.

2015

- Awarded, Executive of the Year Award, Awarded by National Association of Distinguished Professionals
- Awarded, Distinguished Alumni Award, The College of Saint Rose

AREAS OF EXPERTISE

Epidemiology and Infectious Disease (viral, bacteria, air pollution, environmental insurance & exclusion)

Ambient & Indoor Air Quality Modeling

Stationary & Mobile Source Modeling (GHG's, Particulates, Gases, Vapors, Odors)

Asbestos Exposure & Mesothelioma

Lead Assessments and Exposure Pathways

Water Quality Exposures Assessment (Metals, Gases, Particulates)

Silica Dust Monitoring/Analysis/Human Health Assessments

Mold Impacts to Health Exposure & Health Effects

Multi Path Exposure Assessment (Air, Water, & Soil)

Medical Records Data Review for Exposure Matching of Symptoms

Air and Groundwater Health Risk Assessment (TCE, PCE, PCBs, PFOA, PFOS & Environmental Toxins)

Chemical Exposures

Remediation Assessment for Site Contamination to Off-Site Areas

Environmental Zoning and Impact Assessments

Risk Assessments (RCRA, CERCLA)

Environmental Remediation Exposure Assessments/Contamination

Health Risk Toxicology

Product Liability & Technologies

Toxic Tort

Environmental Litigation

Data Validation/Third Party Assessments

Public Health

PLOS ONE

RESEARCH ARTICLE

# Association between Daily Hydrogen Sulfide Exposure and Incidence of Emergency Hospital Visits: A Population-Based Study

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**Data Availability Statement:** According to the permissions of the National Bioethics Committee, the Data Protection Agency, and the Landspitali University Hospital, the data are both ethically and legally restricted. Other researchers who would like to use the data need to apply for permission to the following authorities and institutions: The National Bioethics Committee (vsn@vsn.is), Data Protection Agency (postur@personuvernd.is), and the Landspitali University Hospital (+354 5431000; sidanefnd@landspitali.is). Data on air pollution measurements can be obtained from the

## Abstract

### Background

The adverse health effects of high concentrations of hydrogen sulfide ($H_2S$) exposure are well known, though the possible effects of low concentrations have not been thoroughly studied. The aim was to study short-term associations between modelled ambient low-level concentrations of intermittent hydrogen sulfide ($H_2S$) and emergency hospital visits with heart diseases (HD), respiratory diseases, and stroke as primary diagnosis.

### Methods

The study is population-based, using data from patient-, and population-registers from the only acute care institution in the Reykjavik capital area, between 1 January, 2007 and 30 June, 2014. The study population was individuals (≥18yr) living in the Reykjavik capital area. The $H_2S$ emission originates from a geothermal power plant in the vicinity. A model was used to estimate $H_2S$ exposure in different sections of the area. A generalized linear model assuming Poisson distribution was used to investigate the association between emergency hospital visits and $H_2S$ exposure. Distributed lag models were adjusted for seasonality, gender, age, traffic zones, and other relevant factors. Lag days from 0 to 4 were considered.

### Results

The total number of emergency hospital visits was 32961 with a mean age of 70 years. In fully adjusted un-stratified models, $H_2S$ concentrations exceeding 7.00μg/m$^3$ were associated with increases in emergency hospital visits with HD as primary diagnosis at lag 0 risk ratio (RR): 1.067; 95% confidence interval (CI): 1.024–1.111, lag 2 RR: 1.049; 95%CI:

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1.005–1.095, and lag 4 RR: 1.046; 95%CI: 1.004–1.089. Among males an association was found between $H_2S$ concentrations exceeding 7.00μg/m$^3$, and HD at lag 0 RR: 1.087; 95% CI: 1.032–1.146 and lag 4 RR: 1080; 95%CI: 1.025–1.138; and among those 73 years and older at lag 0 RR: 1.075; 95%CI: 1.014–1.140 and lag 3 RR: 1.072; 95%CI: 1.009–1.139. No associations were found with other diseases.

## Conclusions

The study showed an association between emergency hospital visits with HD as primary diagnosis and same day $H_2S$ concentrations exceeding 7.00μg/m$^3$, more pronounced among males and those 73 years and older than among females and younger individuals.

## Introduction

The adverse health effects of high concentrations of hydrogen sulfide ($H_2S$) exposure are many and relatively well known, as has been reviewed in a report by the World Health Organization [1], but the mechanisms of $H_2S$ toxicity remain debated. Some studies indicate that $H_2S$ inhibit oxygen consumption by mitochondrial oxidase [2], and others suggest that $H_2S$ may affect cysteine residues of most proteins [3]. The first noticeable effect of $H_2S$ is the odour similar to rotten eggs; the odour threshold varies, often considered 7–11 μg/m$^3$ [1,4,5]. With increasing $H_2S$ concentrations other effects appear, for example, eye irritation and neurological symptoms such as headache, nausea, loss of olfactory sense (at 140 mg/m$^3$) [1]. Pulmonary oedema, respiratory arrest, and death may follow a few breaths at 700 mg/m$^3$ [1].

Studies on low-level $H_2S$ exposures have been accumulating through observations of occupational cohorts and populations residing near industries and geothermal fields emitting $H_2S$ and other pollutants [6–13]. These studies have dealt with different outcomes; some have reported association with noticing odour, odour nuisance and decreased daily activity [6,7], increase in respiratory symptoms and anti-asthma drug dispensing [7–9], while others have reported negative associations between long-term $H_2S$ exposure and self-reported asthma and asthma symptoms [10]. Still other studies have reported on neurological symptoms and headaches [8,11,12] while the results on the effect of $H_2S$ exposure on cognitive function remains inconclusive [12,13]. Respiratory mortality and total mortality, as well as lung cancer, have been associated with low-level $H_2S$ exposures [14–16]. Reduced lung function has been reported in two studies [11,17], but was not found in one study [18]. Finally, visits to health care centres and hospitals have been used to study $H_2S$-exposed catchment populations with emphasis on respiratory diseases and cardiovascular diseases where five studies report positive associations [19–22], while a recent study that attempted to evaluate long-term exposure found no association [23].

The comprehensive hospital and population registries operated in Iceland offer a unique opportunity for population-based studies on low-level $H_2S$ exposed inhabitants in the Reykjavik capital area. Since 2006, two geothermal power plants have been located some 30 km east of the city and the characteristic odour of $H_2S$ is occasionally noticed in Reykjavik. The $H_2S$ concentrations have been measured in the capital area with a total population of approximately 196,000 individuals [24].

The aim was to investigate short-term associations between modelled ambient low-level intermittent $H_2S$ concentrations and daily hospital admissions and emergency department

(ED) visits to Landspitali University Hospital (LUH) with heart disease, respiratory disease and stroke as primary diagnoses among individuals living in the Reykjavik capital area.

## Materials and Methods

### Study population

Reykjavik is the world's northernmost capital of a sovereign state and is located in the south-west of Iceland on the southern shore of the Faxafloi bay.

The study period was 1 January 2007 to 30 June 2014. The National Roster, part of the National Registry, kept by Statistics Iceland, was the source of information on the population base (number, age, and gender) which consisted of all individuals, 18 years and older in the Reykjavik capital area. The population was geocoded into sections A to E (see Exposure assessment subchapter) and the total number of individuals in each section (A-E) were calculated within age groups (18-59, 60-72, 73-80, and 81 and older) and gender. The Reykjavik capital area consists of seven municipalities (Alftanes, Gardabaer, Hafnarfjordur, Kopavogur, Mosfellsbaer, Reykjavik, and Seltjarnarnes), defined by community codes and 21 postal codes: 101, 103-105, 107-113, 170, 200, 201, 203, 210, 220, 221, 270, 271, and 276, according to the National Roster 2010 [24].

### Outcome measures

The primary source of data is the records on emergency hospital admissions and ED visits to the only acute care hospital and ED in the Reykjavik capital area at LUH, obtained from the Register of Hospital-treated Patients in Iceland for the study period. Patient data were anonymized and de-identified by LUH specialist prior to data handling. The hospital is operated by the government, and health-care services are financed by taxes. Residents of Iceland are covered by the national health insurance schemes, which pay the bulk of the patients' costs; however, patients pay a certain fee for ambulatory visits. Admission to the hospital is free of charge. The register of Hospital-treated Patients is practically complete, and contains routinely collected data on every patient admission to the hospital and visit to the ED of those 18 years of age or older. Information registered includes the unique registration number of every admission and visit, personal identification numbers according to the National Registry, address, postal code, birth date, gender, admission date, discharge date and discharge diagnoses as diagnosed by the attending physician using the International Classification of Diseases 10th version (ICD-10).

The outcome measure was acute hospital admission, or visit to the ED, reported with one of the following classes of disease: heart disease (HD) (ICD-10 codes: I20-I27: ischaemic heart diseases, I46: cardiac arrest, I48: cardiac arrhythmias, and I50: heart failure), respiratory disease (ICD-10 codes: J20-J22: acute lower respiratory infections, J40-J46: chronic lower respiratory diseases, and J96: respiratory failure) and stroke (ICD-10 codes: I61-I69: cerebrovascular diseases other than I60: subarachnoid haemorrhage and G45: transient cerebral ischaemic attacks and related syndromes and G46: vascular syndromes of brain in cerebrovascular diseases), all as primary diagnosis. The daily number of acute hospital admissions and ED visits were combined and are henceforth referred to as "emergency hospital visits". Encrypted personal identification numbers were used to find individuals with readmissions or revisits within 10 days and with same ICD-10 primary diagnosis; these revisits were excluded, and only the previous admission or ED visit was counted as a visit.

The population was divided into the geographical sections A to E by geocoding the addresses, and the National Roster was used to count the number of people at risk in the sections according to gender and age groups. Patients with an emergency hospital visit were tracked through home

address and geocoded to the exact section A to E, date, and thus assigned $H_2S$ exposure. The parts of the population and patients located outside the borders of section A, and E, were counted with the adjoining sections; see next subchapter and Fig 1.

## Exposure assessment

For the study period 1 January, 2007 to 30 June, 2014, ambient air concentrations and meteorological data were obtained from the Environment Agency of Iceland (EAI), which operates a measurement station located near one of Reykjavik's busiest road intersections (Grensasvegur station, GRE) [25]. The data contained hourly concentration values of nitrogen dioxide ($NO_2$), ozone ($O_3$), particulate matter $\leq$ 10 μm in aerodynamic diameter ($PM_{10}$), sulfur dioxide ($SO_2$) and $H_2S$ measured as micrograms per cubic metre of air ($\mu g/m^3$) as well as hourly values of temperature (°C), relative humidity (RH, %), wind speed (m/s), and wind direction. The devices used to measure pollutant concentrations and calibration frequency have previously been reported [16].

Distance from main roads (>10.000 cars per day) in the Reykjavik capital area was found for each individual's residential street and divided into categories of traffic exposure zones (0-50 m, 51-200 m, 201-500 m, 501-1000 m, and ≥1000 m) and used as a surrogate for traffic-related exposure. Measured air pollution concentrations from the GRE station, were not included in the final analysis as the exposure zones gave a better fit in the final analysis.

The main source of ambient $H_2S$ is from a geothermal power plant located 26 km east of the city centre (Fig 1) [26,27]. Hellisheidi power plant started operation in September 2006. Average $H_2S$ emissions over the study period were 10,532.5 tons annually, fluctuating between 6,902 tons/year in 2007 and 13,340 tons/year in 2010 [28]. Residential distance from the Hellisheidi power plant was adjusted for in the final analysis, by classifying the distance into quartiles (≤22 430 m, 22 431-25 360 m, 25 361-27 330 m, and ≥27 331 m).

To estimate $H_2S$ exposure through 2007 to July 2014 in different sections of the Reykjavik capital area, a simple model was applied whereas the modelled concentration only depends on wind speed, the angle between wind direction and modelled location, and incoming solar radiation. The width of the plume was determined from measurements and calculations using the well known Gaussian plume, Pasquill-Gifford model [29,30], at 25 km from the source under stable conditions [31]. The model predicted $H_2S$ concentrations that were compared to measured concentrations at measurement stations operated by EAI, in section A (Hvaleyrarholt station, HEH) and in section C (GRE) (Fig 1). Emissions from the Nesjavellir power plant were not included in the model, as the power plant is behind a mountain [31], which limits the distribution of $H_2S$ westward in the direction of the Reykjavik capital area [26,27,32], and this was confirmed by $H_2S$ measurement at GRE before the start of the Hellisheidi geothermal power plant in 2006 [16]. The model covers a 50° section from Hellisheidi power plant to the west, which includes the Reykjavik capital area. The concentration was calculated in five 10° sections, defined as A to E (Fig 1). For each section, the average 24-hour $H_2S$ concentration was calculated. The location of Hellisheidi power plant is some 260m above sea level and there is a moderate, practically continuous downward slope [31] westward from the plant to the Reykjavik capital area (GRE). Detailed description of the $H_2S$ modelling can be found in S1 Model Calculations. Model prediction and accuracy was considered sufficient with a Spearman's correlation coefficient of 0.55 for daily averages of $H_2S$ concentrations (Figures D and E in S1 Model Calculations).

Different exposure levels of $H_2S$ were calculated by different percentiles 50% (2.46 $\mu g/m^3$), 60% (3.16 $\mu g/m^3$), 70% (4.14 $\mu g/m^3$), 80% (5.74 $\mu g/m^3$), 85% (7.00 $\mu g/m^3$), 90% (8.80 $\mu g/m^3$) and 95% (11.68 $\mu g/m^3$), and trend analyses were conducted through the percentile levels.

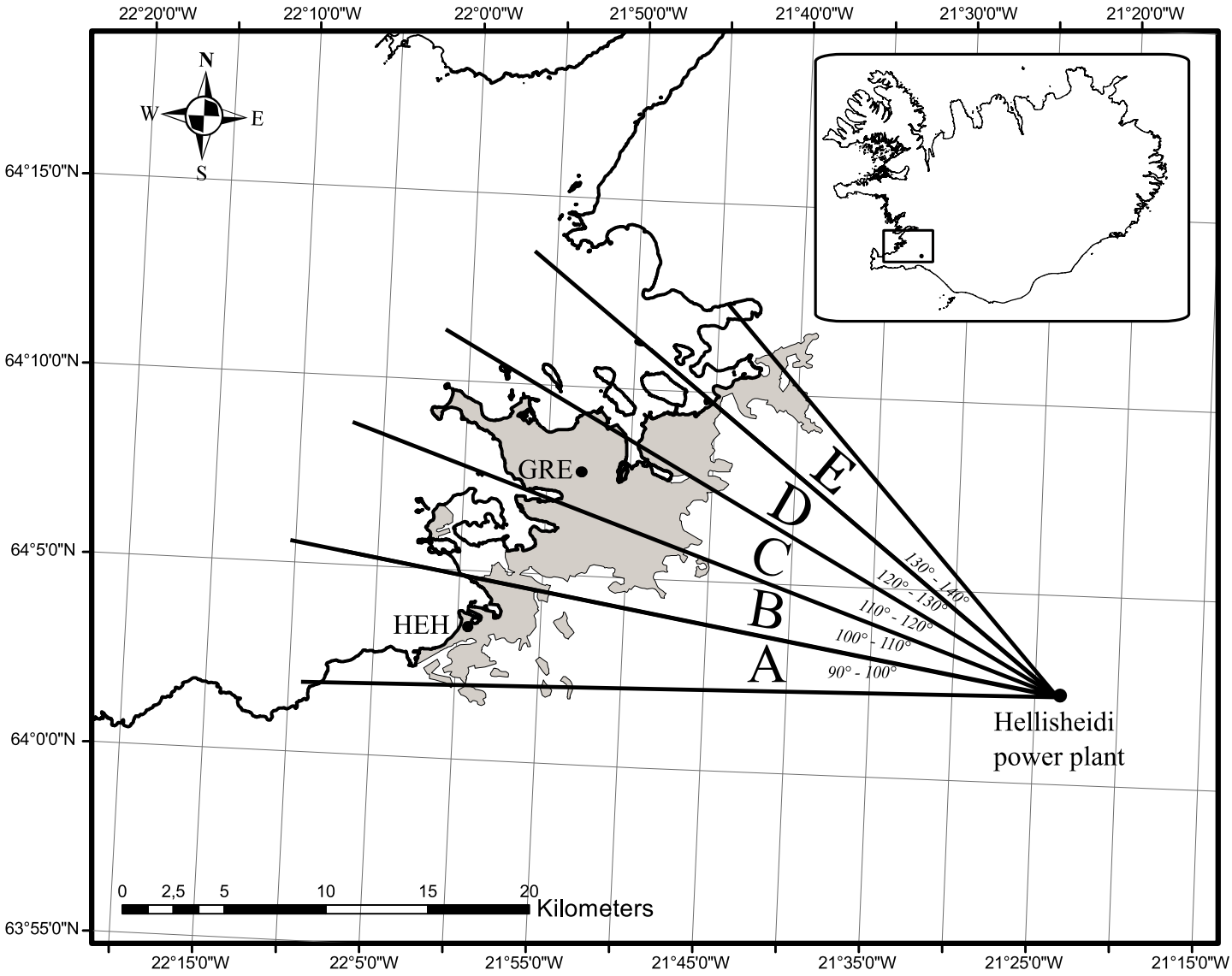


**Fig 1. Five modelled sections (A to E) of the Reykjavik capital area (the shadowed area), and the point source of $H_2S$ emissions, the Hellisheidi power plant.** Small inserted map shows Iceland and the capital's location.

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## Statistical analysis

Daily numbers of emergency hospital visits, with HD, respiratory diseases and stroke as primary diagnoses were counted according to gender and age groups, and time-series plots were made (S1 Fig) as well as time-series plots for $H_2S$ concentrations (Fig 2). We used a generalized linear model (GLM) assuming Poisson distribution of outcome measures to estimate the association between short-term daily exposures to $H_2S$. This method was chosen since hospital admissions and ED visits are a discrete counting event [33] and the method is often used to investigate short-term associations of environmental exposures with various health outcomes [34,35].

Daily numbers of emergency hospital visits were the dependent variable. Separate analyses were performed for HD, respiratory diseases, and stroke as primary diagnosis. Modelled $H_2S$ concentrations at patient's residence were selected as independent variables, classified as

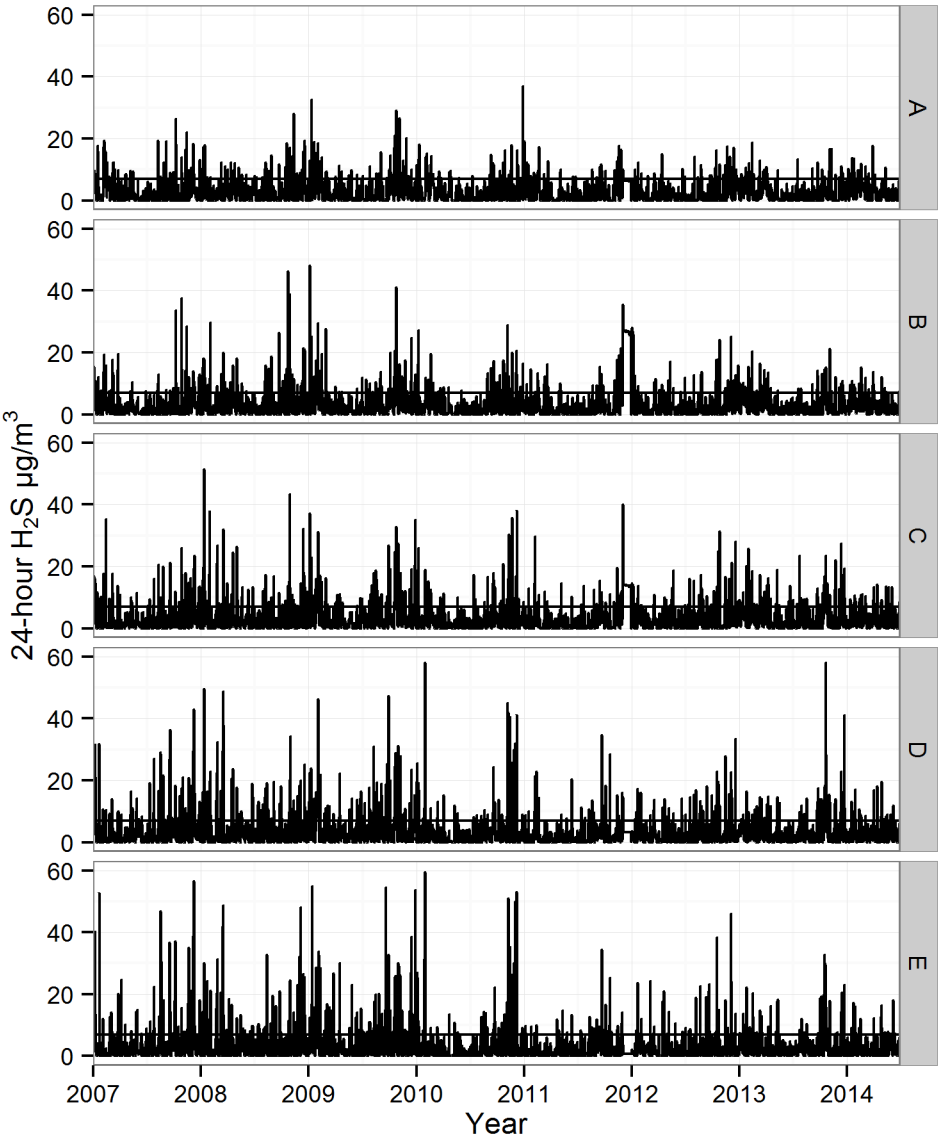


**Fig 2. Daily 24-hour concentrations of $H_2S$ in µg/m$^3$ within modelled sections A to E of the Reykjavik capital area over the study period 1 January, 2007–30 June, 2014.** Horizontal line indicates the 85 percentile limit of 7.00 µg/m$^3$.

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different percentiles of $H_2S$ exposure (50%, 60%, 70%, 80%, 85%, 90% and 95%). Population data was used as offset to account for population size and demographic composition (age and gender) in each section. To control for seasonality and long-term trends in outcome measures, models were adjusted for day-of-week and basic spline with 8 degrees of freedom as it gave the best model fit. The number of degrees of freedom is essential to minimize the autocorrelation in the residuals and to account for seasonal trends in outcome measures [35]. Here, a small number of degrees of freedom was chosen since long-term seasonal trends in number of emergency hospital visits did not seem apparent [34].

A number of models were tested. First, we ran a crude analysis testing the association between $H_2S$ (classified as different percentiles of $H_2S$ exposure) exposure and outcome while adjusting for seasonality (splines) only. Secondly, fully adjusted models were distributed lag

models [35] and were adjusted for seasonality (splines), gender, age group, traffic exposure zone, distance from Hellisheidi power plant, and same-day average temperature using different percentiles of $H_2S$ exposure. Measured concentrations of traffic-related pollution ($NO_2$, $O_3$, $PM_{10}$, and $SO_2$) were tested in the model and did not modify the association, and were thus omitted. Also, potential autocorrelation was avoided by adjusting the model with the number of each outcome measure at lag 1 (previous day). Thirdly, $H_2S$ concentrations at different sections of the Reykjavik capital area were introduced to fully adjusted models as a continuous variable giving results for an increase of 7μg/m$^3$ in $H_2S$ concentrations. Fourthly, dose-response trends were analysed through different percentiles of $H_2S$ exposure levels (50%, 60%, 70%, 80%, 85%, 90%, and 95%) using GLM analysis. Due to dependency of RR estimates within each lag, all $H_2S$ exposure levels were introduced in the model at the same time. Lag days from 0 to 4 were considered in each model. Backwards selection of adjustment variables showed that season, humidity and lags 5 to 7 did not significantly affect the results and were therefore not included in fully adjusted models. Residual analysis and graphical assessment of autocorrelation and spline functions indicated that modelling assumptions were rational.

The analysis yielded risk ratio (RR) and 95% confidence interval (CI) for each lag structure. Here, the focus will be on results for $H_2S$ concentrations exceeding 7.00 μg/m$^3$ (85% exposure level) and emergency hospital visits with HD, respiratory disease, or stroke as primary diagnosis (other results are shown in Supporting Information). Results with p-value less than 0.05 were considered statistically significant.

Data were prepared and statistical analyses were performed using R statistical software, version 3.1.3 [36].

The study and use of the data were approved by Bioethics Committee (VSNb2010120017/03.7), the Data Protection Agency (2010121176AT/), and the Hospital ethics board (Letter dated 2010/12/22).

## Results

The mid-year population of adults (18 years and older) in the Reykjavik capital area was 151095 in year 2010 [24]. During the seven and a half year period (2738 days), there were 13383 patients with a total of 32961 emergency hospital visits to LUH (Table 1), where the proportion of male visits was 56.8%. The average number of daily emergency hospital visits over the study period was 12.0 with a range of 0-32 visits per day (Table 1). Most emergency hospital visits were with HD as primary diagnosis, followed by respiratory diseases. The average number of daily emergency hospital visits with stroke diagnosis was approximately 2.35. Median age of all patients was 73 years. Mean age of patients was 69.9 years with the highest mean age of female HD patients (74.8 years). Patients with respiratory diseases as a primary diagnosis had the youngest mean age (66.5 years). Female patients with emergency hospital visit were on average 3.8 years older than males.

The modelled 24-hour mean concentrations within each section (A-E) are shown in Table 2 and Fig 2. Overall, 75% of all modelled values of 24-hour $H_2S$ concentrations were lower than 5 μg/m$^3$. The mean 24-hour $H_2S$ concentration was highest in section D with an average concentration of 4.04 μg/m$^3$, and lowest in section A (3.02 μg/m$^3$). The highest 24-hour $H_2S$ concentration was 69.5 μg/m$^3$ in section C (Fig 2), and in section A, the highest concentration was 37.0 μg/m$^3$. The number of 24-hour concentrations exceeding the different percentiles and the percentiles' lower limits in μg/m$^3$ within each section are shown in Table 3. The correlation of 24-hour $H_2S$ concentration between sections in the Reykjavik capital area ranged from 0.05 between sections A and E up to 0.80 between sections D and E (Table 2).

**Table 1. Descriptive statistics of daily emergency hospital visits to Landspitali University Hospital, according to primary diagnosis, during 1 January, 2007 to 30 June, 2014.**

| | No. of visits n (%) | Visits/day Mean (±SD) | Visits/day Range | Visits/day Median | Percentiles | |
|---|---|---|---|---|---|---|
| **Emergency hospital patients** | | | | | **25th** | **75th** |
| All primary diagnosis | 32961 (100) | 12.04 (4.86) | 1–32 | 12 | 8 | 15 |
| Females | 14224 (43.2) | 5.28 (2.70) | 0–16 | 5 | 3 | 7 |
| Males | 18737 (56.8) | 6.98 (3.41) | 0–21 | 7 | 4 | 9 |
| Older (≥73yr) | 15885 (48.19) | 5.88 (2.92) | 0–20 | 6 | 4 | 8 |
| Younger (<73yr) | 17076 (51.81) | 6.30 (3.17) | 0–20 | 6 | 4 | 8 |
| Heart diseases as primary diagnosis | 20529 (62.3) | 7.54 (6.65) | 0–23 | 7 | 5 | 10 |
| Females | 7400 | 3.02 (1.77) | 0–12 | 3 | 2 | 4 |
| Males | 13129 | 4.94 (2.72) | 0–19 | 5 | 3 | 7 |
| Older (≥73yr) | 9868 | 3.80 (2,13) | 0–16 | 3 | 2 | 5 |
| Younger (<73yr) | 10661 | 4.11 (2.43) | 0–16 | 4 | 2 | 6 |
| Respiratory diseases as primary diagnosis | 7438 (22.6) | 3.00 (1.80) | 0–13 | 3 | 2 | 4 |
| Females | 4515 | 2.18 (1.32) | 0–10 | 2 | 1 | 3 |
| Males | 2923 | 1.74 (0.98) | 0–7 | 1 | 1 | 2 |
| Older (≥73yr) | 3198 | 1.83 (1.04) | 0–7 | 2 | 1 | 2 |
| Younger (<73yr) | 4240 | 2.12 (1.26) | 0–8 | 2 | 1 | 3 |
| Stroke as primary diagnosis | 4994 (15.2) | 2.35 (1.46) | 0–11 | 2 | 1 | 3 |
| Females | 2309 | 1.65 (0.90) | 0–6 | 1 | 1 | 2 |
| Males | 2685 | 1.81 (1.07) | 0–8 | 1 | 1 | 2 |
| Older (≥73yr) | 2819 | 1.84 (1.06) | 0–9 | 2 | 1 | 2 |
| Younger (<73yr) | 2175 | 1.66 (0.93) | 0–7 | 1 | 1 | 2 |

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**Table 2. Descriptive statistics of modelled daily 24-hour concentrations of $H_2S$ during study period in each section of the Reykjavik capital area, daily count of higher concentration in each section and percentiles, as well as Spearman´s correlation of daily 24-hour concentrations of $H_2S$ between sections.**

| During study period | Section A | Section B | Section C | Section D | Section E |
|---|---|---|---|---|---|
| Modelled days in study period | 2738 | 2738 | 2738 | 2738 | 2738 |
| Mean concentration (μg/m$^3$) (±SD) | 3.02 (4.05) | 3.53 (5.34) | 3.79 (5.96) | 4.04 (6.83) | 3.89 (7.10) |
| Range (μg/m$^3$) | 0–37.0 | 0–48.1 | 0–69.5 | 0–68.2 | 0–66.9 |
| Interquartile range (μg/m$^3$) (0.25, 0.75) | 0.0, 4.8 | 0.1, 4.5 | 0.2, 4.9 | 0.2, 4.9 | 0.2, 4.6 |
| **Number of high concentrations within section** | | | | | |
| **Lower limits of percentiles** | | | | | |
| 50% (≥2.46 μg/m$^3$) | 498 | 986 | 1250 | 862 | 266 |
| 60% (≥3.16 μg/m$^3$) | 436 | 833 | 1091 | 743 | 235 |
| 70% (≥4.14 μg/m$^3$) | 345 | 632 | 903 | 567 | 188 |
| 80% (≥5.74 μg/m$^3$) | 241 | 461 | 646 | 393 | 135 |
| 85% (≥7.00 μg/m$^3$) | 177 | 362 | 533 | 321 | 106 |
| 90% (≥8.80 μg/m$^3$) | 124 | 257 | 395 | 263 | 75 |
| 95% (≥11.68 μg/m$^3$) | 58 | 158 | 259 | 171 | 49 |
| **Spearman´s correlation** | | | | | |
| Section A | 1.00 | | | | |
| Section B | 0.67 | 1.00 | | | |
| Section C | 0.39 | 0.73 | 1.00 | | |
| Section D | 0.17 | 0.37 | 0.75 | 1.00 | |
| Section E | 0.05 | 0.18 | 0.46 | 0.80 | 1.00 |

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**Table 3. Number of emergency hospital visits to Landspitali University Hospital, in each modelled section of the Reykjavik capital area, and in higher percentiles of $H_2S$ concentrations during 1 January, 2007 to 30 June, 2014.**

| Lower limits of percentiles | All sections | Section A | Section B | Section C | Section D | Section E |
|---|---|---|---|---|---|---|
| 50% (≥2.46 μg/m$^3$) | 14157 | 835 | 2813 | 8130 | 1985 | 394 |
| 60% (≥3.16 μg/m$^3$) | 12137 | 730 | 2381 | 6961 | 1714 | 351 |
| 70% (≥4.14 μg/m$^3$) | 9614 | 585 | 1802 | 5669 | 1284 | 274 |
| 80% (≥5.74 μg/m$^3$) | 6853 | 397 | 1340 | 4031 | 878 | 207 |
| 85% (≥7.00 μg/m$^3$) | 5596 | 288 | 1067 | 3347 | 726 | 168 |
| 90% (≥8.80 μg/m$^3$) | 4089 | 199 | 746 | 2453 | 578 | 113 |
| 95% (≥11.68 μg/m$^3$) | 2616 | 93 | 463 | 1604 | 381 | 75 |
| Total visits (%) | 32961 (100) | 1895 (5.7) | 6678 (20.3) | 18934 (57.4) | 4502 (13.7) | 952 (2.9) |
| Total inhabitants (%) | 151095 (100) | 11868 (7.9) | 29168 (19.3) | 83703 (55.4) | 20220 (13.4) | 6136 (4.1) |

doi:10.1371/journal.pone.0154946.t003

The crude analysis for the association of $H_2S$ concentrations exceeding 7.00 μg/m$^3$ and HD, respiratory diseases and stroke as primary diagnosis when adjusting only for seasonality (splines) is shown in S1 Table. An overall increase in RRs was seen for every outcome measure at every lag, though the confidence intervals (CI) were wide and included unity. In S2 Table shows the RR for every 7.00 μg/m$^3$ increase in $H_2S$ concentration (introduced as continuous variable) for HD diseases, respiratory diseases, and stroke as primary diagnosis from fully adjusted models. The only CI not including unity was for stroke as primary diagnosis at lag 2.

In the fully adjusted analysis in un-stratified models, statistically significant associations were found between $H_2S$ concentrations exceeding 7.00 μg/m$^3$ and increases in emergency hospital visits with HD as primary diagnosis at lags 0, 2, and 4. There were also increases at lags 1 and, 3, though CIs included unity (Fig 3). Trend analyses between different levels of exposure (from 50 to 95 percentiles) gave $p<0.05$ at lags 0 and 2, indicating a positive dose-response association (Table 4). P-values for trend analysis at lag 4 were also $<0.05$ for a negative dose-response (Table 4). When analysis was stratified by gender, associations were found among males and between $H_2S$ concentrations exceeding 7.00 μg/m$^3$ and HD at lags 0 and 4 (Fig 3); however the trend analysis between different exposure levels was not significant at any lag. Among females, an association was found at lag 3 (Fig 3). Also among females, p-values for trend analysis between different exposure levels indicated a positive dose-response association at lags 0 and 2 but a negative dose-response at lag 4 (Table 4). Analyses stratified by age showed associations between $H_2S$ concentrations exceeding 7.00 μg/m$^3$ among those 73 years and older at lags 0 and 3, whereas CIs did not include unity (Table 4). Additionally, p-values for trend analysis were $<0.05$ at lags 0, 2, and 3, indicating a positive dose-response association (Table 4).

The RRs for the association between $H_2S$ at different percentiles and emergency hospital visits with respiratory diseases as primary diagnosis is shown in Table 5. In fully adjusted analysis, both un-stratified and stratified by gender and age, models for $H_2S$ concentrations exceeding 7.00 μg/m$^3$ were not statistically associated with an increase or decrease in emergency hospital visits with respiratory diagnosis at any lag (Table 5). On the other hand, some trends through different levels of exposure (from 50 to 95 percentiles) were significant at lag 0, and two other lags in the un-stratified analysis, and in male and the older strata, indicating a negative dose-response association (Table 5).

The RRs for the association between $H_2S$ at different percentiles and emergency hospital visits with stroke as primary diagnosis are shown in Table 6. In the fully adjusted analysis in un-stratified models, non-significant associations between $H_2S$ concentrations exceeding 7.00 μg/m$^3$ and emergency hospital visits with stroke were found (Table 6). When analysis was

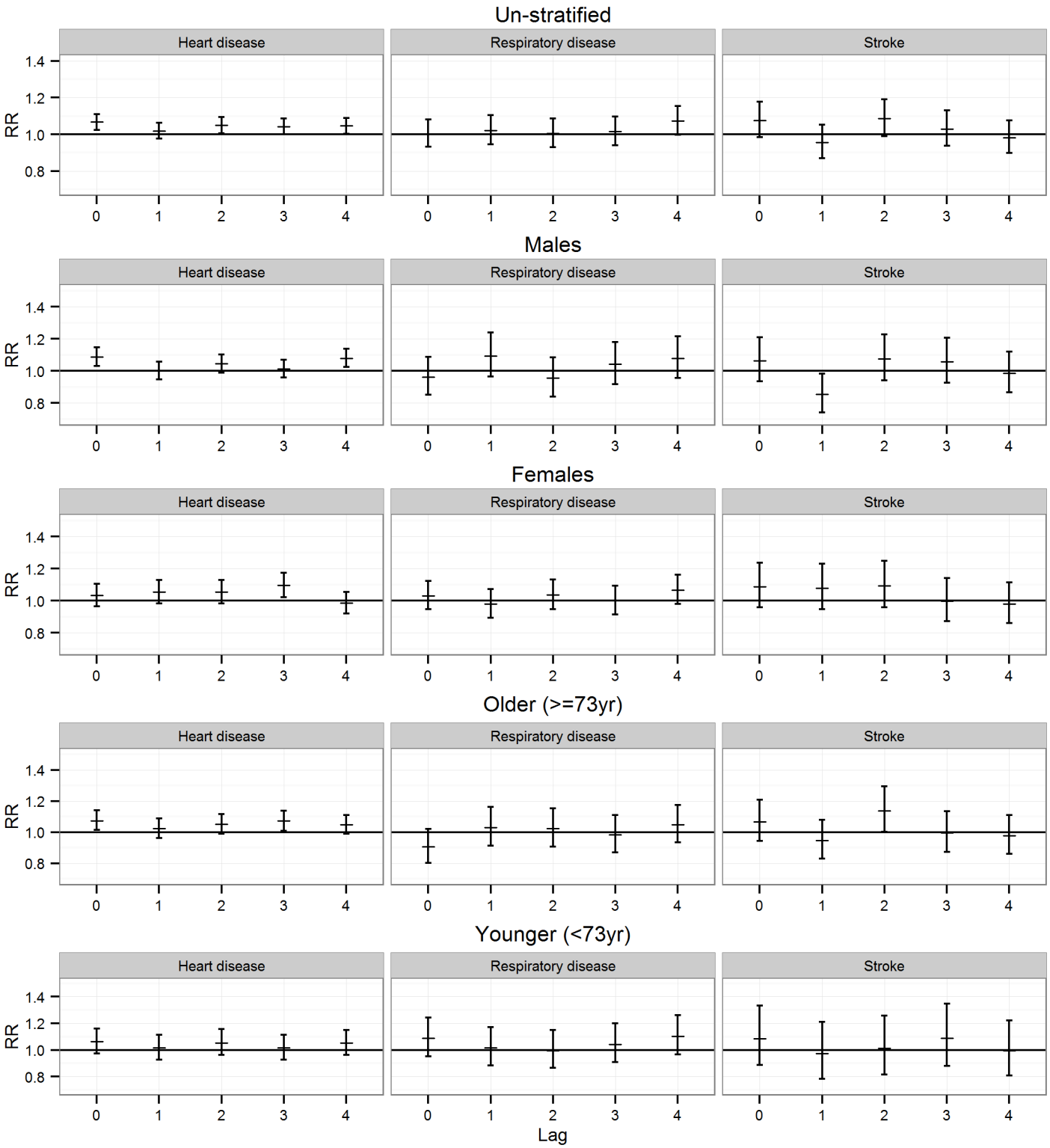


**Fig 3. Associations between daily emergency hospital visits with heart diseases, respiratory diseases, and stroke as primary diagnosis and $H_2S$ concentrations exceeding 7.00 μg/m$^3$ in fully adjusted models for lags 0–4, un-stratified, and gender and age stratification.**

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**Table 4. Associations between daily emergency hospital visits with heart diseases as primary diagnosis and different percentiles of $H_2S$ exposure in fully adjusted models for lags 0–4, un-stratified, and gender and age stratification.**

| | 50% (≥2.46 μg/m³) | | 60% (≥3.16 μg/m³) | | 70% (≥4.14 μg/m³) | | 80% (≥5.74 μg/m³) | | 85% (≥7.00 μg/m³) | | 90% (≥8.80 μg/m³) | | 95% (≥11.68 μg/m³) | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Lag** | **RR** | **95% CI** | **RR** | **95% CI** | **RR** | **95% CI** | **RR** | **95% CI** | **RR** | **95% CI** | **RR** | **95% CI** | **RR** | **95% CI** | **p-trend** |
| **Un-stratified**[a] | | | | | | | | | | | | | | | |
| **0** | 1.007 | 1.004, 1.009 | 1.056 | 1.023, 1.091 | 1.048 | 1.013, 1.084 | 1.068 | 1.028, 1.109 | 1.067 | 1.024, 1.111 | 1.071 | 1.022, 1.121 | 1.059 | 1.001, 1.122 | 0.0038 |
| **1** | 1.008 | 1.005, 1.010 | 1.073 | 1.037, 1.109 | 1.056 | 1.019, 1.094 | 1.031 | 0.991, 1.072 | 1.019 | 0.976, 1.064 | 1.011 | 0.962, 1.061 | 0.986 | 0.928, 1.049 | 0.1927 |
| **2** | 1.006 | 1.003, 1.009 | 1.043 | 1.009, 1.079 | 1.047 | 1.011, 1.085 | 1.057 | 1.017, 1.100 | 1.049 | 1.005, 1.095 | 1.045 | 0.995, 1.098 | 1.062 | 1.001, 1.128 | 0.0027 |
| **3** | 1.005 | 1.003, 1.008 | 1.053 | 1.019, 1.089 | 1.043 | 1.007, 1.081 | 1.034 | 0.994, 1.075 | 1.042 | 0.999, 1.087 | 1.012 | 0.964, 1.063 | 1.010 | 0.952, 1.072 | 0.7116 |
| **4** | 1.060 | 1.027, 1.094 | 1.050 | 1.016, 1.084 | 1.043 | 1.008, 1.079 | 1.037 | 0.998, 1.077 | 1.046 | 1.004, 1.089 | 1.053 | 1.005, 1.103 | 1.020 | 0.962, 1.081 | 0.0483 |
| **Gender stratification**[b] | | | | | | | | | | | | | | | |
| **Males** | | | | | | | | | | | | | | | |
| **0** | 1.010 | 1.006, 1.013 | 1.078 | 1.034, 1.124 | 1.072 | 1.026, 1.121 | 1.091 | 1.039, 1.145 | 1.087 | 1.032, 1.146 | 1.080 | 1.018, 1.146 | 1.067 | 0.992, 1.148 | 0.0627 |
| **1** | 1.007 | 1.003, 1.010 | 1.066 | 1.021, 1.113 | 1.061 | 1.014, 1.111 | 1.023 | 0.973, 1.077 | 1.000 | 0.946, 1.057 | 1.017 | 0.955, 1.083 | 1.017 | 0.940, 1.100 | 0.4006 |
| **2** | 1.006 | 1.003, 1.009 | 1.052 | 1.007, 1.099 | 1.043 | 0.996, 1.092 | 1.053 | 1.001, 1.107 | 1.045 | 0.989, 1.104 | 1.042 | 0.978, 1.110 | 1.033 | 0.955, 1.116 | 0.3038 |
| **3** | 1.005 | 1.002, 1.008 | 1.034 | 0.991, 1.080 | 1.028 | 0.982, 1.076 | 1.018 | 0.968, 1.071 | 1.012 | 0.958, 1.069 | 0.978 | 0.918, 1.042 | 0.983 | 0.910, 1.062 | 0.0879 |
| **4** | 1.085 | 1.042, 1.130 | 1.070 | 1.027, 1.116 | 1.057 | 1.012, 1.105 | 1.066 | 1.015, 1.119 | 1.080 | 1.025, 1.138 | 1.089 | 1.026, 1.156 | 1.065 | 0.989, 1.147 | 0.9065 |
| **Females** | | | | | | | | | | | | | | | |
| **0** | 1.002 | 0.998, 1.006 | 1.019 | 0.967, 1.075 | 1.006 | 0.951, 1.064 | 1.029 | 0.967, 1.095 | 1.033 | 0.965, 1.105 | 1.055 | 0.978, 1.139 | 1.047 | 0.953, 1.151 | 0.0000 |
| **1** | 1.010 | 1.005, 1.014 | 1.087 | 1.029, 1.148 | 1.046 | 0.987, 1.109 | 1.045 | 0.980, 1.114 | 1.054 | 0.983, 1.130 | 1.000 | 0.923, 1.084 | 0.933 | 0.843, 1.033 | 0.1910 |
| **2** | 1.006 | 1.002, 1.010 | 1.025 | 0.970, 1.083 | 1.052 | 0.993, 1.115 | 1.061 | 0.996, 1.131 | 1.054 | 0.983, 1.129 | 1.047 | 0.967, 1.134 | 1.113 | 1.011, 1.225 | 0.0004 |
| **3** | 1.006 | 1.001, 1.010 | 1.086 | 1.029, 1.147 | 1.069 | 1.009, 1.132 | 1.060 | 0.995, 1.130 | 1.096 | 1.023, 1.174 | 1.076 | 0.994, 1.163 | 1.060 | 0.963, 1.166 | 0.1761 |
| **4** | 1.015 | 0.964, 1.069 | 1.013 | 0.960, 1.068 | 1.018 | 0.963, 1.077 | 0.987 | 0.928, 1.050 | 0.985 | 0.921, 1.054 | 0.987 | 0.914, 1.066 | 0.940 | 0.854, 1.034 | 0.0010 |
| **Age stratification**[c] | | | | | | | | | | | | | | | |
| **Older (≥73yr)** | | | | | | | | | | | | | | | |
| **0** | 1.008 | 1.004, 1.011 | 1.062 | 1.014, 1.113 | 1.059 | 1.008, 1.112 | 1.069 | 1.012, 1.129 | 1.075 | 1.014, 1.140 | 1.080 | 1.011, 1.154 | 1.096 | 1.010, 1.189 | 0.0000 |
| **1** | 1.010 | 1.006, 1.014 | 1.093 | 1.042, 1.147 | 1.053 | 1.001, 1.109 | 1.037 | 0.980, 1.097 | 1.024 | 0.963, 1.090 | 1.026 | 0.956, 1.100 | 0.993 | 0.909, 1.085 | 0.2710 |
| **2** | 1.006 | 1.003, 1.010 | 1.023 | 0.975, 1.074 | 1.048 | 0.996, 1.103 | 1.056 | 0.998, 1.117 | 1.052 | 0.990, 1.118 | 1.045 | 0.974, 1.121 | 1.057 | 0.97, 1.152 | 0.0000 |
| **3** | 1.007 | 1.003, 1.011 | 1.058 | 1.008, 1.110 | 1.042 | 0.991, 1.096 | 1.061 | 1.003, 1.122 | 1.072 | 1.009, 1.139 | 1.077 | 1.006, 1.154 | 1.080 | 0.993, 1.174 | 0.0000 |
| **4** | 1.036 | 0.99, 1.0840 | 1.049 | 1.001, 1.099 | 1.054 | 1.004, 1.107 | 1.034 | 0.979, 1.091 | 1.049 | 0.989, 1.112 | 1.069 | 1.000, 1.142 | 1.006 | 0.926, 1.093 | 0.7273 |
| **Younger (<73yr)** | | | | | | | | | | | | | | | |
| **0** | 1.007 | 1.001, 1.012 | 1.056 | 0.985, 1.133 | 1.042 | 0.968, 1.122 | 1.073 | 0.989, 1.163 | 1.062 | 0.973, 1.160 | 1.064 | 0.963, 1.175 | 1.026 | 0.906, 1.162 | 0.2849 |
| **1** | 1.006 | 1.000, 1.012 | 1.056 | 0.982, 1.135 | 1.064 | 0.986, 1.148 | 1.026 | 0.943, 1.117 | 1.016 | 0.927, 1.114 | 1.002 | 0.902, 1.113 | 0.988 | 0.866, 1.127 | 0.2173 |
| **2** | 1.006 | 1.000, 1.012 | 1.070 | 0.996, 1.151 | 1.050 | 0.973, 1.134 | 1.061 | 0.976, 1.154 | 1.054 | 0.962, 1.155 | 1.047 | 0.943, 1.163 | 1.076 | 0.946, 1.224 | 0.0926 |
| **3** | 1.004 | 0.999, 1.010 | 1.059 | 0.985, 1.138 | 1.052 | 0.975, 1.135 | 1.011 | 0.929, 1.100 | 1.017 | 0.928, 1.115 | 0.953 | 0.856, 1.060 | 0.949 | 0.833, 1.081 | 0.0541 |
| **4** | 1.084 | 1.013, 1.161 | 1.049 | 0.978, 1.125 | 1.030 | 0.957, 1.108 | 1.049 | 0.968, 1.138 | 1.051 | 0.962, 1.148 | 1.052 | 0.950, 1.164 | 1.047 | 0.924, 1.186 | 0.2137 |

[a]Un-stratified models were adjusted for gender, age group, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

[b]Gender stratified models were adjusted for age group, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

[c]Age stratified models were adjusted for gender, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature

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**Table 5. Associations between daily emergency hospital visits with respiratory diseases as primary diagnosis and different percentiles of $H_2S$ exposure in fully adjusted models for lags 0–4, un-stratified, and gender and age stratification.**

| | 50% (≥2.46 μg/m³) | | 60% (≥3.16 μg/m³) | | 70% (≥4.14 μg/m³) | | 80% (≥5.74 μg/m³) | | 85% (≥7.00 μg/m³) | | 90% (≥8.80 μg/m³) | | 95% (≥11.68 μg/m³) | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | p-trend |
| **Un-stratified**[a] | | | | | | | | | | | | | | | |
| **0** | 1.009 | 1.005, 1.014 | 1.059 | 0.999, 1.123 | 1.040 | 0.978, 1.106 | 1.012 | 0.945, 1.083 | 1.003 | 0.931, 1.081 | 0.979 | 0.899, 1.065 | 0.980 | 0.882, 1.088 | 0.0340 |
| **1** | 1.004 | 0.999, 1.009 | 1.038 | 0.977, 1.103 | 1.011 | 0.948, 1.078 | 1.031 | 0.961, 1.107 | 1.022 | 0.946, 1.105 | 1.031 | 0.944, 1.126 | 1.003 | 0.898, 1.121 | 0.9040 |
| **2** | 1.005 | 1.001, 1.010 | 1.024 | 0.964, 1.088 | 1.045 | 0.980, 1.114 | 1.047 | 0.976, 1.123 | 1.005 | 0.930, 1.086 | 0.973 | 0.890, 1.064 | 0.951 | 0.851, 1.063 | 0.1702 |
| **3** | 1.006 | 1.002, 1.011 | 1.055 | 0.993, 1.120 | 1.026 | 0.962, 1.093 | 1.004 | 0.936, 1.077 | 1.015 | 0.940, 1.096 | 1.001 | 0.917, 1.093 | 0.950 | 0.851, 1.059 | 0.0721 |
| **4** | 1.101 | 1.040, 1.165 | 1.105 | 1.043, 1.172 | 1.096 | 1.031, 1.164 | 1.108 | 1.037, 1.185 | 1.073 | 0.998, 1.155 | 1.085 | 0.999, 1.179 | 1.106 | 0.999, 1.225 | 0.4642 |
| **Gender stratification**[b] | | | | | | | | | | | | | | | |
| **Males** | | | | | | | | | | | | | | | |
| **0** | 1.007 | 0.999, 1.014 | 1.032 | 0.938, 1.136 | 1.026 | 0.928, 1.135 | 0.988 | 0.882, 1.105 | 0.963 | 0.851, 1.089 | 0.950 | 0.825, 1.093 | 0.944 | 0.793, 1.123 | 0.0003 |
| **1** | 1.000 | 0.993, 1.008 | 1.053 | 0.953, 1.163 | 1.015 | 0.914, 1.128 | 1.075 | 0.958, 1.207 | 1.094 | 0.965, 1.240 | 1.066 | 0.923, 1.230 | 1.021 | 0.851, 1.224 | 0.2308 |
| **2** | 1.006 | 0.999, 1.014 | 1.040 | 0.942, 1.149 | 1.028 | 0.926, 1.142 | 0.999 | 0.889, 1.123 | 0.955 | 0.841, 1.085 | 0.939 | 0.811, 1.089 | 0.922 | 0.768, 1.108 | 0.0009 |
| **3** | 1.006 | 0.999, 1.014 | 1.046 | 0.948, 1.154 | 1.039 | 0.937, 1.153 | 1.000 | 0.891, 1.123 | 1.042 | 0.919, 1.181 | 1.002 | 0.867, 1.158 | 0.971 | 0.813, 1.160 | 0.2599 |
| **4** | 1.112 | 1.013, 1.221 | 1.143 | 1.039, 1.256 | 1.138 | 1.031, 1.257 | 1.152 | 1.033, 1.284 | 1.078 | 0.956, 1.216 | 1.065 | 0.928, 1.221 | 1.132 | 0.959, 1.336 | 0.4502 |
| **Females** | | | | | | | | | | | | | | | |
| **0** | 1.011 | 1.005, 1.016 | 1.079 | 1.008, 1.154 | 1.051 | 0.978, 1.128 | 1.028 | 0.950, 1.113 | 1.031 | 0.946, 1.124 | 0.996 | 0.903, 1.098 | 1.003 | 0.888, 1.132 | 0.2065 |
| **1** | 1.006 | 1.001, 1.012 | 1.028 | 0.958, 1.103 | 1.009 | 0.936, 1.087 | 1.003 | 0.923, 1.089 | 0.980 | 0.895, 1.073 | 1.012 | 0.914, 1.122 | 0.994 | 0.874, 1.131 | 0.1771 |
| **2** | 1.005 | 1.000, 1.010 | 1.014 | 0.945, 1.089 | 1.056 | 0.980, 1.138 | 1.077 | 0.993, 1.169 | 1.037 | 0.948, 1.134 | 0.991 | 0.894, 1.099 | 0.967 | 0.850, 1.099 | 0.6453 |
| **3** | 1.006 | 1.001, 1.012 | 1.061 | 0.990, 1.138 | 1.017 | 0.944, 1.095 | 1.007 | 0.928, 1.093 | 1.001 | 0.915, 1.095 | 1.004 | 0.907, 1.112 | 0.936 | 0.824, 1.064 | 0.0572 |
| **4** | 1.093 | 1.023, 1.168 | 1.080 | 1.010, 1.156 | 1.066 | 0.993, 1.145 | 1.079 | 0.998, 1.166 | 1.067 | 0.980, 1.162 | 1.098 | 0.997, 1.208 | 1.090 | 0.968, 1.228 | 0.8976 |
| **Age stratification**[c] | | | | | | | | | | | | | | | |
| **Older (≥73yr)** | | | | | | | | | | | | | | | |
| **0** | 1.003 | 0.996, 1.010 | 0.971 | 0.885, 1.064 | 0.934 | 0.847, 1.030 | 0.916 | 0.821, 1.022 | 0.906 | 0.804, 1.022 | 0.862 | 0.751, 0.990 | 0.868 | 0.732, 1.029 | 0.0000 |
| **1** | 1.005 | 0.998, 1.012 | 1.098 | 0.999, 1.207 | 1.035 | 0.936, 1.145 | 1.050 | 0.940, 1.173 | 1.029 | 0.912, 1.163 | 1.059 | 0.922, 1.215 | 1.014 | 0.852, 1.207 | 0.8427 |
| **2** | 1.005 | 0.998, 1.013 | 1.003 | 0.912, 1.103 | 1.055 | 0.954, 1.166 | 1.057 | 0.946, 1.180 | 1.023 | 0.906, 1.154 | 0.994 | 0.864, 1.143 | 0.983 | 0.827, 1.168 | 0.7260 |
| **3** | 1.006 | 0.999, 1.013 | 1.037 | 0.943, 1.139 | 1.014 | 0.918, 1.120 | 0.974 | 0.872, 1.088 | 0.983 | 0.871, 1.109 | 0.894 | 0.776, 1.030 | 0.895 | 0.753, 1.064 | 0.0013 |
| **4** | 1.059 | 0.969, 1.158 | 1.063 | 0.970, 1.164 | 1.029 | 0.934, 1.133 | 1.064 | 0.957, 1.183 | 1.048 | 0.934, 1.176 | 1.106 | 0.971, 1.259 | 1.131 | 0.966, 1.325 | 0.0881 |
| **Younger (<73yr)** | | | | | | | | | | | | | | | |
| **0** | 1.015 | 1.006, 1.023 | 1.143 | 1.028, 1.271 | 1.137 | 1.017, 1.271 | 1.093 | 0.966, 1.237 | 1.087 | 0.951, 1.243 | 1.079 | 0.927, 1.255 | 1.079 | 0.895, 1.300 | 0.8132 |
| **1** | 1.004 | 0.995, 1.013 | 0.992 | 0.888, 1.109 | 0.994 | 0.884, 1.118 | 1.019 | 0.895, 1.160 | 1.018 | 0.884, 1.172 | 1.010 | 0.860, 1.187 | 0.990 | 0.809, 1.212 | 0.6274 |
| **2** | 1.006 | 0.998, 1.015 | 1.046 | 0.936, 1.169 | 1.042 | 0.927, 1.172 | 1.041 | 0.915, 1.184 | 0.996 | 0.865, 1.148 | 0.960 | 0.815, 1.131 | 0.932 | 0.759, 1.143 | 0.0543 |
| **3** | 1.007 | 0.998, 1.015 | 1.075 | 0.963, 1.201 | 1.039 | 0.925, 1.168 | 1.034 | 0.909, 1.176 | 1.043 | 0.907, 1.200 | 1.094 | 0.934, 1.281 | 0.995 | 0.816, 1.213 | 0.8912 |
| **4** | 1.138 | 1.025, 1.264 | 1.144 | 1.029, 1.272 | 1.152 | 1.031, 1.287 | 1.151 | 1.019, 1.299 | 1.103 | 0.966, 1.261 | 1.081 | 0.929, 1.258 | 1.099 | 0.912, 1.326 | 0.0274 |

[a]Models adjusted for gender, age group, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

[b]Models adjusted for age group, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

[c]Models adjusted for gender, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

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**Table 6. Association between daily emergency hospital visits with stroke as primary diagnosis and different percentiles of $H_2S$ exposure in fully adjusted models for lags 0–4, un-stratified, and gender and age stratification.**

| | 50% (≥2.46 μg/m³) | | 60% (≥3.16 μg/m³) | | 70% (≥4.14 μg/m³) | | 80% (≥5.74 μg/m³) | | 85% (≥7.00 μg/m³) | | 90% (≥8.80 μg/m³) | | 95% (≥11.68 μg/m³) | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | RR | 95% CI | p-trend |
| **Un-stratified**[a] | | | | | | | | | | | | | | | |
| **0** | 1.010 | 1.005, 1.016 | 1.074 | 1.000, 1.153 | 1.058 | 0.981, 1.140 | 1.049 | 0.965, 1.140 | 1.076 | 0.984, 1.178 | 1.081 | 0.977, 1.197 | 1.126 | 0.996, 1.274 | 0.0038 |
| **1** | 1.004 | 0.998, 1.009 | 1.016 | 0.943, 1.093 | 1.008 | 0.932, 1.089 | 0.966 | 0.885, 1.054 | 0.957 | 0.870, 1.053 | 0.956 | 0.858, 1.066 | 0.983 | 0.860, 1.124 | 0.0086 |
| **2** | 1.009 | 1.003, 1.014 | 1.109 | 1.030, 1.193 | 1.170 | 1.083, 1.263 | 1.130 | 1.038, 1.231 | 1.086 | 0.989, 1.192 | 1.145 | 1.031, 1.272 | 1.105 | 0.970, 1.258 | 0.2032 |
| **3** | 1.007 | 1.001, 1.013 | 1.036 | 0.963, 1.115 | 1.004 | 0.929, 1.085 | 1.044 | 0.958, 1.137 | 1.029 | 0.937, 1.130 | 1.019 | 0.916, 1.135 | 0.984 | 0.861, 1.124 | 0.7005 |
| **4** | 1.018 | 0.950, 1.092 | 1.029 | 0.958, 1.104 | 1.036 | 0.961, 1.116 | 0.987 | 0.908, 1.073 | 0.983 | 0.897, 1.077 | 0.950 | 0.855, 1.056 | 0.894 | 0.783, 1.021 | 0.0013 |
| **Gender stratification**[b] | | | | | | | | | | | | | | | |
| **Males** | | | | | | | | | | | | | | | |
| **0** | 1.011 | 1.003, 1.018 | 1.069 | 0.967, 1.182 | 1.030 | 0.926, 1.145 | 1.031 | 0.915, 1.160 | 1.065 | 0.937, 1.211 | 1.105 | 0.957, 1.275 | 1.152 | 0.968, 1.372 | 0.0104 |
| **1** | 0.999 | 0.991, 1.007 | 1.000 | 0.901, 1.110 | 1.001 | 0.896, 1.118 | 0.899 | 0.792, 1.019 | 0.854 | 0.743, 0.982 | 0.860 | 0.735, 1.008 | 0.898 | 0.738, 1.092 | 0.0002 |
| **2** | 1.008 | 1.000, 1.016 | 1.071 | 0.966, 1.189 | 1.114 | 0.998, 1.242 | 1.101 | 0.976, 1.243 | 1.076 | 0.942, 1.228 | 1.158 | 0.997, 1.344 | 1.076 | 0.892, 1.298 | 0.0654 |
| **3** | 1.009 | 1.001, 1.017 | 1.068 | 0.963, 1.185 | 1.022 | 0.916, 1.141 | 1.072 | 0.950, 1.209 | 1.058 | 0.928, 1.207 | 1.039 | 0.893, 1.209 | 1.016 | 0.841, 1.227 | 0.7996 |
| **4** | 1.021 | 0.926, 1.127 | 1.004 | 0.908, 1.111 | 1.045 | 0.940, 1.161 | 0.995 | 0.884, 1.119 | 0.985 | 0.866, 1.121 | 0.886 | 0.761, 1.031 | 0.789 | 0.648, 0.961 | 0.0148 |
| **Females** | | | | | | | | | | | | | | | |
| **0** | 1.010 | 1.002, 1.018 | 1.079 | 0.976, 1.194 | 1.091 | 0.981, 1.213 | 1.071 | 0.953, 1.204 | 1.089 | 0.960, 1.236 | 1.054 | 0.913, 1.218 | 1.098 | 0.922, 1.307 | 0.1021 |
| **1** | 1.008 | 1.000, 1.017 | 1.035 | 0.932, 1.149 | 1.016 | 0.910, 1.134 | 1.043 | 0.924, 1.178 | 1.079 | 0.946, 1.231 | 1.071 | 0.923, 1.244 | 1.080 | 0.898, 1.299 | 0.0000 |
| **2** | 1.009 | 1.001, 1.017 | 1.152 | 1.038, 1.278 | 1.234 | 1.107, 1.376 | 1.162 | 1.031, 1.309 | 1.095 | 0.960, 1.248 | 1.129 | 0.974, 1.310 | 1.134 | 0.946, 1.358 | 0.4811 |
| **3** | 1.005 | 0.997, 1.013 | 1.002 | 0.902, 1.112 | 0.984 | 0.881, 1.098 | 1.014 | 0.897, 1.145 | 0.998 | 0.873, 1.141 | 0.997 | 0.856, 1.161 | 0.949 | 0.787, 1.145 | 0.1973 |
| **4** | 1.014 | 0.918, 1.120 | 1.055 | 0.954, 1.167 | 1.024 | 0.921, 1.139 | 0.977 | 0.868, 1.101 | 0.980 | 0.860, 1.116 | 1.025 | 0.885, 1.186 | 1.016 | 0.849, 1.216 | 0.3792 |
| **Age stratification**[c] | | | | | | | | | | | | | | | |
| **Older (≥73yr)** | | | | | | | | | | | | | | | |
| **0** | 1.010 | 1.002, 1.017 | 1.068 | 0.968, 1.178 | 1.047 | 0.944, 1.162 | 1.038 | 0.925, 1.165 | 1.068 | 0.943, 1.210 | 1.076 | 0.935, 1.238 | 1.151 | 0.972, 1.363 | 0.0136 |
| **1** | 1.007 | 0.999, 1.015 | 1.031 | 0.931, 1.141 | 0.984 | 0.883, 1.096 | 0.966 | 0.857, 1.091 | 0.947 | 0.829, 1.081 | 0.963 | 0.829, 1.118 | 0.975 | 0.811, 1.173 | 0.0042 |
| **2** | 1.011 | 1.003, 1.019 | 1.107 | 1.000, 1.225 | 1.228 | 1.105, 1.365 | 1.165 | 1.036, 1.310 | 1.138 | 1.002, 1.294 | 1.191 | 1.032, 1.376 | 1.242 | 1.043, 1.479 | 0.0059 |
| **3** | 1.006 | 0.998, 1.013 | 1.025 | 0.926, 1.135 | 0.982 | 0.882, 1.093 | 1.031 | 0.915, 1.161 | 0.996 | 0.874, 1.135 | 1.000 | 0.862, 1.161 | 0.899 | 0.745, 1.084 | 0.1683 |
| **4** | 1.030 | 0.935, 1.135 | 1.037 | 0.940, 1.144 | 1.056 | 0.952, 1.171 | 0.952 | 0.847, 1.070 | 0.978 | 0.861, 1.110 | 0.949 | 0.820, 1.098 | 0.905 | 0.753, 1.088 | 0.0005 |
| **Younger (<73yr)** | | | | | | | | | | | | | | | |
| **0** | 1.011 | 0.999, 1.024 | 1.092 | 0.928, 1.284 | 1.085 | 0.914, 1.286 | 1.068 | 0.883, 1.291 | 1.086 | 0.885, 1.333 | 1.090 | 0.864, 1.375 | 1.088 | 0.817, 1.448 | 0.0591 |
| **1** | 0.999 | 0.986, 1.013 | 0.998 | 0.843, 1.183 | 1.048 | 0.877, 1.252 | 0.962 | 0.787, 1.175 | 0.973 | 0.782, 1.211 | 0.953 | 0.742, 1.223 | 1.014 | 0.743, 1.382 | 0.4534 |
| **2** | 1.005 | 0.992, 1.018 | 1.115 | 0.942, 1.320 | 1.085 | 0.908, 1.296 | 1.074 | 0.883, 1.308 | 1.012 | 0.815, 1.257 | 1.082 | 0.848, 1.381 | 0.911 | 0.665, 1.246 | 0.3775 |
| **3** | 1.010 | 0.997, 1.024 | 1.061 | 0.897, 1.256 | 1.044 | 0.875, 1.247 | 1.077 | 0.886, 1.309 | 1.089 | 0.880, 1.347 | 1.050 | 0.822, 1.341 | 1.140 | 0.848, 1.533 | 0.0064 |
| **4** | 1.005 | 0.858, 1.178 | 1.015 | 0.862, 1.195 | 1.006 | 0.847, 1.194 | 1.044 | 0.865, 1.261 | 0.993 | 0.806, 1.223 | 0.963 | 0.757, 1.225 | 0.891 | 0.658, 1.208 | 0.0996 |

[a]Models adjusted for gender, age group, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

[b]Models adjusted for age group, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature.

[c]Models adjusted for gender, season, day-of-week, distance from Hellisheidi power plant, traffic exposure zone and temperature

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stratified by age, a statistically significant association was found at lag 2 among those 73 years and older (Fig 3, and in Table 6). In the trend analyses through different levels of exposure (from 50 to 95 percentiles), statistically significant positive association was found at lag 0 in un-stratified, males, and the older stratum, and statistically significant negative association at lag 1 in the same stratum, indicating dose-response manner of association (Table 6).

## Discussion

The present study showed an association between hospital admission and emergency department visits with heart disease (HD) as primary diagnosis and $H_2S$ concentrations exceeding 7.00 μg/m$^3$ occurring the same day, more pronounced among males and those 73 years and older than among females and younger individuals. The associations were also seen with delay of two and four days. Same-day associations were gradually increasing through higher percentiles of exposure in a dose-response manner of relationship. These results are in concordance with previous studies on possible short-term effects, which were based on hospital records [19–22]. The present results are also supported by a study on association of increased general mortality and $H_2S$ exposure, conducted in the Reykjavik capital area [16], and by another study in the same setting on increased dispensing of anti-asthma drugs and $H_2S$ levels [9]. On the other hand, in the medical literature, it has been discussed that low-level $H_2S$ intracellularly may reduce vasoconstriction and promote cardiovascular health [37,38]. Contrary, $H_2S$ exposure in healthy human volunteers as low as 5 ppm were associated with a shift from aerobic to anaerobic metabolism [39], however, whether this has relevance in the present study is merely speculative.

The chosen time unit in the present study for $H_2S$ concentrations and for emergency hospital visits was 24 hours. The argument for the definition of the 24-hour $H_2S$ μg/m$^3$ was that the distribution was skewed, as shown in Table 2 and Fig 2 and in S2 and S3 Figs, where only one or two times per month did the $H_2S$ concentration exceed the odour limit of 7.00 $H_2S$ μg/m$^3$ over two consecutive days. The rationale for the definition of sum of admissions and visits per 24 hours (and not for example hourly admissions and visits) is the diurnal distribution, where hospital visits accumulate during office hours (shown in S4 Fig).

In the present study, the pattern seen in increased risk of emergency hospital visits associated with increased percentiles of $H_2S$ concentrations is compatible with a harvesting effect [40]; namely, there is a positive dose-response association at lag 0 and lag 2, and negative dose-response association at lag 4 through the un-stratified analysis, and among females. The hypothesized harvesting effect [40] is not very obvious from the lag analyses. However, in the calculation on $H_2S$ concentrations exceeding 7.00 μg/m$^3$, the risk estimation was highest for lag 0 and lowest for lag 1 in un-stratified, male stratum, and both age strata, giving further support for harvesting effect and the possibility of short-term effects of $H_2S$ exposure. Considering the intermittent $H_2S$ exposure, however, it is not possible to evaluate long-term health effects of $H_2S$ exposure in the present study setting, and for that a reference group from an unexposed region may be needed. The outcome in the present study, emergency hospital visits with HD as primary diagnosis, brings to mind the many known risks and causal factors for these diseases. However, respecting other causal effects, these should not preclude the possible short-term effect of an environmental pollution such as $H_2S$. The present results are not in contradiction with the studies from Rotorua using a cross-sectional approach, which have not found associations between long-term $H_2S$ exposure and asthma and chronic obstructive pulmonary diseases [10,23]. Here, emergency hospital visits with respiratory diseases were not associated with $H_2S$ exposure; however, the study had limited power to detect such possible association.

Backwards selection showed that the traffic exposure zone (distance from main roads) was a better fit in the fully adjusted analysis rather than the measured traffic-related pollution ($NO_2$, $O_3$, $PM_{10}$, and $SO_2$) from one measurement station in Reykjavik (GRE).

## Strengths

To our best knowledge, the Reykjavik capital area population is the largest population so far that has been investigated concerning possible adverse health effect of $H_2S$ exposure where hospital data is used as an outcome. The comprehensive hospital and population registries and the general use of personal identification numbers also strengthen the study, as they allowed us to eliminate readmission and revisits to an acceptable level. The National Roster was used to obtain the population data in each section as well as for the information on location of the patients within the sections attending the hospital, LUH, thus derived from the same source. As the LUH is the only health care institution in the Reykjavik capital area offering acute hospital and emergency department service to the population in the area, there is no competition from other similar health care institutions. However, services are also provided by general practitioners and medical specialists in out-of-hospital offices.

The estimation of the $H_2S$ exposure in the different sections of the Reykjavik capital area was done by simple model, as the main variables of wind direction and speed, and solar radiation are routinely measured by a governmental institution EAI independently from the continuous accumulation of the outcome information at the LUH. The model estimated $H_2S$ concentrations in five different sections of the city, which is an improvement from exposure estimates from a single monitoring station in the Reykjavik area, as it gives a more individual-based estimation of exposure. The width of the plume and importance of the distance from the source (the inhabited zones in Reykjavik capital area range 20 to 30 km from source) were taken into account, and have been discussed previously [31].

The methods used in the present study follow the well known, widely accepted, and documented approaches [35].

## Limitations

The exposure data is derived from a simple model for the $H_2S$ exposure applied in five sections of the Reykjavik capital area, instead of containing data on individual exposure. Also, it is known, that individuals are exposed to air pollution in various other places than their home. This was nonetheless not taken into account whereas information on the patient's residence was only available data, so some misclassification of $H_2S$ exposure is possible. Additionally, misclassification of calculated $H_2S$ exposure is plausible but should be minimal as the difference between the measurements at GRE and the model calculations is small, helped by the fact that nearly zero values occur when the wind direction is not from the east. However, this approach using modelled $H_2S$ estimations is an advance from the use of concentration measurements obtained from only one measurement station in the Reykjavik capital, as has been used in previous studies [9,16]. Also, we are adjusting for residential distance from main traffic roads as a surrogate for other daily levels of individually measured airborne pollutants, which improved the model fit and gave a better adjustment than measured concentrations from one measurement station (GRE).

In the present study, we were not able to adjust for social variables, or premorbid condition of those with emergency hospital visits. The relative risk sizes found are very small and, even if statistically significant because of the large sample size, they could possibly be accounted for by a small amount of unknown confounding. Detailed investigation of the role of these important factors awaits future studies.

Our study is conducted on material originating from a single academic health care institution in a capital area, which may limit the generalizability of the results; however, the hospital and the ED are together the only institution of their kind serving the catchment population as a community hospital, rendering the study population-based. The characteristics of the population are known and the population is relatively homogenous, being 95-99% white Caucasian [24], and there is a uniform financing of the health-care and insurance.

We tested several approaches in the analyses of the association of the $H_2S$ exposure and the many components of the outcome measures in an attempt to yield as much knowledge as possible from our data set. The high number of calculations performed may give rise to concern due to multiple comparison problems; however, it has been argued that no adjustments are needed for these [41].

The increased risk for emergency hospital visits with HD as primary diagnosis seemed to be marked for those 73 years of age and older and the population data was restricted to those 18 years and older. This limits the generalizability of the results with regard to age. Another limitation is the relatively small number of cases with diagnoses of respiratory diseases and stroke, rendering analyses of these outcomes statistically underpowered. This is an inherent weakness for studies in small populations. The counting of admissions and visits to LUH were restricted to attendances where HD, respiratory diseases, and stroke were a primary diagnosis of the individuals; thus we were not able to analyse all attendances to LUH. The quality of the routine medical diagnoses at LUH has not been evaluated in a separate study, a weakness that this study shares with most other studies relying on hospital records. Finally, according to the diurnal distribution of the admissions and visits in the study, it was not realistic to achieve a narrower time frame than 24 hours in the association analysis.

## Conclusions

The results from this study indicate an increase in hospital admission and emergency department visits with heart disease as primary diagnosis associated with $H_2S$ concentrations exceeding 7.00 $\mu g/m^3$ the same day, more pronounced among males and among those 73 years and older. The associations were also seen with delay of two or four days. The same-day associations were gradually increasing through higher percentiles of exposure in a dose-response manner. These results were adjusted for gender, age, season, traffic-related pollution, and number of lags with generally accepted and acknowledged methods. These results are further supported by a previous study in the same setting showing increased general mortality when 24-hour $H_2S$ concentrations exceed 7.00 $\mu g/m^3$.

## Supporting Information

**S1 Fig. Daily number of emergency hospital visits with heart diseases, respiratory diseases, and stroke as primary diagnosis in the Reykjavik capital area over the study period 1 January, 2007 to 30 June, 2014.**
(TIFF)

**S2 Fig. Daily 24-hour concentrations of $H_2S$ in $\mu g/m^3$ within modelled sections A to E of the Reykjavik capital area in 2009.** The horizontal line indicates the 85 percentile limit of 7.00 $\mu g/m^3$.
(TIFF)

**S3 Fig. Daily 24-hour concentrations of $H_2S$ in $\mu g/m^3$ within sections A to E of the Reykjavik capital area in November 2009.** The horizontal line indicates the 85 percentile lower

limit 7.00 μg/$m^3$.
(TIFF)

**S4 Fig. Hourly number of emergency hospital visits with heart diseases, respiratory diseases, and stroke as primary diagnosis in the Reykjavik capital area during 2009.**
(TIFF)

**S1 Model Calculations. Description of hydrogen sulfide concentrations modelling for five different sections of the Reykjavik capital area of Iceland.**
(PDF)

**S1 Table. Crude results for associations between daily emergency hospital visits with heart diseases, respiratory diseases and stroke as primary diagnosis and $H_2S$ concentrations exceeding 7.00 μg/$m^3$ for lags 0-4 (adjusted for seasonality only).**
(DOCX)

**S2 Table. Associations between daily emergency hospital visits with heart diseases, respiratory diseases, and stroke as primary diagnosis for 7.00 μg/$m^3$ changes in $H_2S$ concentrations (introduced as continuous variable) in fully adjusted models for lags 0-4.**
(DOCX)

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## Author Contributions

Conceived and designed the experiments: RGF HKC TT AO SHL TG VR. Performed the experiments: RGF SHL. Analyzed the data: RGF SHL HKC AO. Contributed reagents/materials/analysis tools: TT. Wrote the paper: RGF VR. Revision of study for important intellectual content: RGF HKC TT AO SHL TG VR. Revision of manuscript: RGF HKC TT AO SHL TG VR.

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# Air pollution impacts from a pulp and paper mill facility located in adjacent communities, Edmundston, New Brunswick, Canada and Madawaska, Maine, United States

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United States toxics release inventory (US TRI)

ABSTRACT

Pulp and paper mills are known large emitters of air pollution creating potential environmental challenges and human health impacts. This study assesses air pollution and associated impacts from connected pulp and paper facilities which operate in Edmundston, New Brunswick, Canada, and Madawaska, Maine, United States (US). Despite operating within two regulatory jurisdictions on both sides of the international border (in Canada and the US), the mills have exceeded their approvals to operate and national air quality regulations multiple times between 2010 and 2017. Previous studies have linked many types of cancer as well as respiratory and cardiovascular diseases to air pollution emitted by pulp and paper mills, making this an important regional issue in Canada and the US. While potential environmental and community impacts are likely similar on both sides of the international border, emissions from each side are not reported on the other, and few studies have been conducted on either side. Historic and ongoing lack of local epidemiological health and environmental impact studies and recommendations for improvements to mill operations to improve environmental and human health are presented.

## 1. Introduction

Pulp and paper mills are known for being large emitters of air pollution (Cheremisinoff and Rosenfeld, 2010). The pulp and paper mills which operate out of Edmundston, New Brunswick (NB) in Canada and Madawaska, Maine (ME) in the United States (US), respectively, are better known for their relatively unique cross-border operations than any environmental damages they may cause. The pulp and paper facilities are divided by country, with pulp facilities in Canada and paper facilities in the US but are both operated by the Twin Rivers Paper Company (Fig. 1). The two facilities are connected via pipelines bringing pulp and steam from Edmundston to Madawaska (Twin Rivers Paper Company, 2019; Twin Rivers Paper Company, 2019). Production capacities are cited as 370,000 tons per year of bleached magnesium sulphite and groundwood pulp from Edmundston and 335,000 tons per year of paper from Madawaska (Twin Rivers Paper Company, 2019).

Pulp and paper mills around the world have been found to exceed regulatory guidelines on air pollution (*e.g.*, Hoffman et al. 2017). In Canada and the US, air pollutant emissions data from pulp and paper mills are generally self-monitored and self-reported to the federal governments by the facilities (Johnston Edwards and Walker, 2019). Fines may be imposed for exceedances of air quality regulations but are not mandatory (e.g., Hoffman et al., 2015).

Air pollution impacts in combination from the mills in Edmundston and Madawaska have not been reported in scholarly literature, despite their connected nature. A couple of scholarly reports include health impacts from the mill in Edmundston (Milewski and Liu, 2009; Milewski, 2012) but fail to consider that air pollutants can easily cross the international border. No scholarly reports were found on the impact of air pollutants emitted by the Madawaska mill. In this paper, we focus on air pollution impacts from the two mill operations in Edmundston and Madawaska: the current state of pollutant exceedances, regulations, management, and mitigation. We will also make

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**Fig. 1.** Map showing locations of the mills in Edmundston, NB and Madawaska, ME (GoogleEarth, 2020).

recommendations on how these might be improved for the two connected facilities.

### 1.1. Historical context

NB and ME are both known for having many pulp and paper operations in the twentieth century, though operations have slowed down or ceased entirely in many locations since mid-century (Deprez et al., 1986). Both areas were largely forested, which resulted in a plethora of logging and logging-related industries. In the case of the pulp and paper facilities in Edmundston and Madawaska, the current operator Twin Rivers Paper Company cites their operational history to have begun with the 1906 opening of a lumber mill in Plaster Rock, NB, followed shortly by the 1916 opening of the Edmundston pulp facility (Twin Rivers Paper Company, 2019). The Madawaska paper facility opened in 1925 with two paper machines using pulp piped across the river, and rapidly increased in capacity (Twin Rivers Paper Company, 2019). Several modernizations were carried out in the latter half of the twentieth century, though the previous owners, Fraser Papers, were renamed to Twin Rivers Paper Company in 2010 and the mills were purchased in 2013 (Twin Rivers Paper Company, 2019).

### 1.2. Contaminants of concern

Many air pollutants are emitted by pulp and paper mill operations (*e.g.*, Hoffman et al., 2015). Fine particulate matter (PM) is considered a pollutant. Other pollutants due to their specific effects on human and ecosystem health, include nitrogen oxides ($NO_x$), sulphur oxides ($SO_x$), lead, volatile organic compounds (VOCs), and carbon monoxide (CO). The Canadian National Pollutant Release Inventory (NPRI) is a government program with policy objectives around identifying and reducing pollution levels which collects data on many air contaminants from companies with greater than ten employees (Johnston Edwards and Walker, 2019). Based on the publicly available NPRI data for 2017, other substances released into the air by the pulp mill in Edmundston include: ammonia, arsenic, cadmium, chlorine, chlorine dioxide ($ClO_2$), cumene, dioxins and furans, formaldehyde, hexavalent chromium, isopropyl alcohol, manganese, mercury, methanol, methyl ethyl ketone, polycyclic aromatic hydrocarbons (PAHs), phosphorus, selenium, sulphuric acid, and zinc (Environment and Climate Change Canada, 2018). The paper mill similarly reports its annual releases of lead, mercury, polycyclic aromatic compounds, and benzo (g,h,i) perylene to the United States Toxics Release Inventory (US TRI) (United States Environmental Protection Agency, 2019). The US TRI was established in 1987 by the US EPA and is a compilation of facility-reported information about certain chemicals (*i.e.*, those that cause cancer or chronic human health problems, or significant acute human or environmental problems) that gets reported to the public (Johnston Edwards and Walker, 2019; United States Environmental Protection Agency, 2020). Many of these compounds are known carcinogens, including arsenic, cadmium, dioxins and furans, and formaldehyde, with still others being probably or possibly carcinogenic (United States Environmental Protection Agency, 2018).

## 2. Human health and environmental impacts

### 2.1. Human health impacts

Air pollution can negatively impact human health (*e.g.*, Anenberg et al., 2018; Chen et al., 2020; Paul et al., 2020). For example, common concerns due to the contaminants listed above

**Table 1**
Specific cancers and diseases for which increased risk has been shown to be due to working at a pulp and/or paper mill, and which studies have found each.

| Increased risk of disease or increased mortality from | Statistically Significant? | Type of mill process examined | Reference |
|---|---|---|---|
| Large intestine cancer | Yes | Sulphate | Sala-Serra et al. (1996) |
| Breast cancer | No | Sulphate | Sala-Serra et al. (1996) |
| Malignant neoplasms | No | Sulphate | (in women) (Sala-Serra et al., 1996) |
| Esophagus cancer | Yes | Exposed to both, but excess likely due to sulphite | Band et al. (1997) |
| Pleura cancer | Yes | Both | Band et al. (1997) |
| Kidney cancer | Yes | Both | Band et al. (1997) |
| Brain cancer | Yes | Both | Band et al. (1997) |
| Gall bladder cancer | Yes | Both | Band et al. (1997) |
| Non-Hodgkin's lymphoma | Yes | Both | Band et al. (1997) |
| Hodgkin's disease | Yes | Sulphite | Band et al. (1997) |
| All cancers | Yes | Unspecified | McLean et al. (2006) |
| Lung cancer | Yes | Both | (in women) (Andersson et al., 2009) |
| Lung cancer | Yes | Paper | Torén et al. (1996) |
| Lung cancer | Yes | Both | (linked to $SO_2$ exposure) (Lee et al. (2002) |
| Cardiovascular disease | Mostly no | Both | (in men) (Jäppinen and Tola, 1990) |
| Cardiovascular disease | Yes, in some departments | Both | (in women) (Langseth and Kjaerheim, 2006) |
| Circulatory system disease | Only in sulphate mills | Sulphite, sulphate, and paper | (in men) (Jäppinen, 1987) |
| Stomach cancer | No | Sulphite | Rix et al. (1997) |
| Stomach cancer | Yes | Sulphite | Torén et al. (1996) |
| Pancreatic cancer | No | Sulphite | Rix et al. (1997) |
| Testicular cancer | Only for sulphate | Both | Andersson et al. (2012) |
| Lymphosarcoma and reticulum cell sarcoma | Yes | Sulphite | Torén et al. (1996) |
| Chronic bronchitis | Yes | Both | (linked to $SO_2$ and other irritant gases) (Andersson et al. 2013) |

include cancer and cardiovascular disease. Unfortunately, no peer-reviewed epidemiological studies have been conducted on pulp and paper workers or the general community in Edmundston (Milewski and Liu, 2009). However, studies have been conducted on workers at other pulp and paper mills that use similar processes (*e.g.*, Deprez et al., 1986; Jäppinen, 1987; Torén et al., 1996; Andersson et al., 2009) and in communities near other pulp and paper facilities (*e.g.*, Haahtela et al., 1992; Marttila et al., 1994; Jaakkola et al., 1999; Mirabelli and Wing, 2006; Soskolne and Sieswerda, 2010).

There is broad-based recognition of negative health consequences due to working in pulp and paper mills across many countries, though those cannot be solely attributed to air pollution (*e.g.*, Jäppinen, 1987; Torén et al., 1996; Andersson et al., 2009; Sala-Serra et al., 1996; Band et al., 1997; Rix et al., 1997; Lee et al., 2002; McLean et al., 2006; Langseth and Kjaerheim; 2006, Andersson et al., 2012). Table 1 lists cancers and diseases for which increased risk has been shown due to working at pulp or paper mills as an illustration of the types of health effects, though it is unlikely that the surrounding population experiences all of these, due to their reduced exposure to mill-based contaminants in comparison to workers. Despite a plethora of health problems associated with workers' exposure at pulp or paper facilities, many studies debate which health issues can be directly attributed to mill-based exposure as opposed to confounding factors like smoking or natural causes. Some studies do not differentiate between smokers and non-smokers, which has been postulated to be a confounding factor in certain health impacts (e.g., lung cancer) (Rix et al., 1997). Similarly, other studies do not always differentiate between workers in sulphate and sulphite mills, but diseases linked only with sulphite mills have been listed here when possible (*e.g.*, Band et al., 1997), as the Edmundston mill uses a combination of sulphite and mechanical processes for pulping. Several studies included in this assessment have examined effects of select atmospheric pollutants rather than overall effects of working in a pulp or paper mill, including the examination of volatile organochlorine exposure by McLean et al. (2006) and the investigation of paper dust by Langseth and Kjaerheim (Langseth and Kjaerheim, 2006). Sulphur dioxide ($SO_2$) exposure in pulp and paper mills across 12 countries, including both kraft and sulphite mills, was found to possibly be related to increased risk of lung cancer (Lee et al., 2002). In Finnish sulphite mills, $SO_2$ exposure was also linked to excess cardiovascular deaths in men (Jäppinen and Tola, 1990). $SO_2$, chlorine, and $ClO_2$ gases were all found to increase risk of chronic bronchitis with exposure at high concentrations in Swedish pulp mill workers, roughly two-thirds of which worked in sulphite mills (Andersson et al., 2013).

While there has been comparatively little research conducted on health impacts of air pollution due to pulp and paper mills at a community level, most existing research suggests that negative health impacts do exist. For example, Jaakkola et al., (1999) showed that exposed populations had relative changes of -0.57 mean respiratory infections per person-year, along with -4.1% with cough symptoms and -6.4% with headache or migraine in the previous year corresponding to a reduction of mean total reduced sulphur from 11 $\mu g/m^3$ to 6 $\mu g/m^3$ measured over the respective preceding years. Even small concentrations of pollutants might increase respiratory and eye irritation in children (Marttila et al., 1994). Further, proximity to a pulp mill was a relevant factor in higher rates of daytime wheezing in adolescents exposed to tobacco smoke (Mirabelli and Wing, 2006). Not all community-based research has found health impacts due to the proximity of a mill. Deprez et al. (1986) found that for the 66 ME towns studied located near pulp and paper mills in 1980–1982, home location did not appear to influence hospital admission rates for respiratory diseases. However, the towns were selected as having a town centre within 24 km of a mill, and so did not consider possible variations due to population density, relatively large town area, or prevailing winds (Deprez et al., 1986). A study by Milewski and Liu (2009) reported on the incidence of cancer in various NB communities. However, the study did not differentiate possible causes of cancers in Edmundston, making it difficult to measure impacts attributed solely from the mill. Further, most data were from 1989-2005, which may not reflect current cancer rates or current impacts from air pollution in the area.

The magnitude and extent of air pollution impacts on environmental and human health from the pulp and paper mills in Edmundston and Madawaska are largely unknown, though some could be inferred from the NPRI and US TRI self-reported data. However, there are insufficient studies about the human health effects of air pollution from

the mills on the local communities, and existing studies fail to examine variables such as distance from the mills, predominant wind conditions, or events during which pollutants exceeded legal limits. Indeed, this is generally true of air pollution studies from pulp and paper mills in the region (*e.g.*, Hoffman et al., 2017; Hoffman et al., 2015). The only studies we found about human health impacts did not discuss the mills in any detail beyond stating their presence as a possible contributor to cancer rates in the region via production of pollutants listed in the NPRI (Milewski, 2012) or listing the mills as risk factors and stating that no health risk assessments had been done in the area (Milewski and Liu, 2009). Neither Milewski (2012) or Milewski and Liu (2009) appears to have considered pollutants reported to the US TRI, though both mills are quite proximate to the international border.

### 2.2. Environmental impacts

Studies in other areas have shown that air pollution can negatively impact lichen chemistry composition and lichen biodiversity (Walker et al., 2006; Walker et al., 2009). Reduced lichen biodiversity has been shown to be an effective proxy for presence of air pollution, with pollution intolerant species being negatively affected to varying levels by different pollutants (*e.g.*, (Perlmutter, 2010; Malaspina et al., 2018). Various species of lichens have been found to be sensitive to atmospheric $SO_2$, ammonia, and other pollutants (*e.g.*, Walker et al., 2003, Paoli et al., 2015). Das et al. (2013) used lichens to map air pollution around a paper mill, finding that lichen diversity generally increased with increasing distance from the mill, although other pollution sources and predominant wind directions may have confounded results. Lichen biodiversity around Edmundston and Madawaska could be used as a monitoring tool to examine the extent and magnitude, if any, of air pollution impacts arising from the mills.

### 2.3. Monitoring programs

Both Canada and the United States have laws limiting the amount of air pollutants that can be emitted per unit time. NB regulates air pollution via the *Clean Air Act*, under which the pulp mill is considered a "Class 1 facility," requiring an approval to operate (Government of New Brunswick, 2016). The approval lays out quantities of pollutants that the mill is allowed to legally emit, the monitoring required by the province, and reports that must be filed about emissions (Government of New Brunswick, 2018).

Due to the cross-border nature of both facilities, the pulp and paper mills report separately to their different jurisdictions about the air pollutants that they emit. The Edmundston pulp mill reports all annual releases to the NPRI (Environment and Climate Change Canada, 2018), as mandated by its formation under section 48, "National inventory," of the *Canadian Environmental Protection Act, 1999*. The Madawaska paper mill, similarly, reports air pollutant emissions to the Annual Air Emissions Inventory (AAEI) in ME (Air Emissions Inventory Data, 2019). It also reports some annual releases to the federal US TRI (United States Environmental Protection Agency, 2019). Unfortunately, effects of air pollution cannot be constrained by political borders, and the NPRI does not include pollutant releases from the Madawaska mill. Therefore, we combined Canadian and US inventories to calculate the total effect of both facilities on air pollution (Table 2). The paper facility in ME emits fewer major air pollutants into the environment compared to the pulp facility in NB over both 2016 and 2017.

There are many limitations in the current monitoring strategies for air pollution emitted by the facilities. Both the NPRI and the AAEI are self-reported by Twin Rivers, which casts doubt on the validity of the data. Further, there are few regional air quality stations nearby, and they mainly focus on $PM_{2.5}$ and $SO_2$ (New Brunswick Department of Environment and Local Government, 2016), so it is difficult to estimate the zone of elevated contaminants due to the mill emissions and thus the zone of possible impacts to human and environmental health. The Edmundston meteorological station is located roughly 5 km north of the mills, but predominant wind directions are such that pollutants from the mills are relatively infrequently carried toward instruments at this location (Fig. S1). We suspect that this is part of why $NO_2$, $PM_{2.5}$, $O_3$, and $SO_2$ measured at this location do not exceed 1 h limits (Figs. S2, S3, S4, and S5, respectively), but $SO_2$ measurements nearer the mills do exceed legal limits and are generally higher (Fig. S6). $PM_{2.5}$ measurements nearer the mills also have much higher peaks than at the meteorological station (Figs. S7 and S3, respectively). The Canadian National Air Pollution Surveillance (NAPS) program information would generally be included in these comparisons as a comparison to the facility-reported data, but there is no data for Edmundston in 2016, and data is only available for 12% of the year in 2017 (National Air Surveillance Program, 2021).

## 3. Regulation

As the pulp and paper mills have cross-border operations, both the US and Canada are involved in regulation. In NB, the Twin Rivers Paper Company Inc. is currently subject to Approval to Operate I-10081, pursuant to the Air Quality Regulation of the NB *Clean Air Act*. This approval outlines emergency reporting, non-emergency reporting, $SO_2$ and total PM emissions caps, specific limits (see Table 3) and monitors for various machines, and general reporting of annual and monthly air quality required for operation (Government of New Brunswick, 2018). The approval is valid for five years (2018–2023), and replaced a similar approval to operate, labelled I-8229, which was valid for the previous five years (Government of New Brunswick, 2018). Minor offences are penalized by monetary fines, but it appears that not all exceedances are penalized, given the relative number of exceedances listed by the company (New Brunswick Department of Environment and Local Government, 2018) and the fines listed by the province (Government of New Brunswick, 2016).

The approval to operate at the Edmundston pulp mill is very similar to that of another pulp mill in NB, located in Atholville. While the Atholville mill emissions limits are generally lower, it is a bleached dissolving grade sulphite pulp with production of 400 air-dry metric tonnes per day (Government of New Brunswick, 2015), while the Edmundston mill has capacity of 700 air dry tonnes per day of bleached sulphite and nominal production capacity of 360 air dry tonnes per day of bleached/unbleached groundwood, which is substantially larger (Government of New Brunswick, 2018). Table 3 shows differences in regulated air pollutant emissions; for completion, the Madawaska paper mill is also included.

In ME, the company is subject to its Title V Permit; this permit clarifies what the facility needs to do to control its output air pollution to meet the US *Clean Air Act* regulations (United States Environmental Protection Agency, 2017). The ME Department of Environmental Protection assesses the changes, decides on best practical treatment of contaminants (including reasonably available control technology), determines what monitoring and compliance reports must occur, and generally sets forth how the company will act in accordance with the *Clean Air Act* for the next five years before a renewal of the Title V Permit is required (Department of Environmental Protection, 2018). Changes to operations in the mill must be reported during this period, with minor and major amendments issued in the interim (Department of Environmental Protection, 2019; Department of Environmental Protection, 2019).

### 3.1. Contaminant thresholds and exceedances

ME and NB have similar but not identical thresholds for many contaminants. The *Clean Air Act* in NB lays out thresholds for common contaminants as concentrations not to be exceeded per unit time. As such, CO, hydrogen sulphide ($H_2S$), nitrogen dioxide ($NO_2$), $SO_2$, and total suspended particulate all have maximum concentrations over various averaging time periods, *e.g.*, 1, 8, 24 h, 1 year (New Brunswick Department of Environment and Local Government, 2002). The *Clean Air Act*

**Table 2**
Combined air emissions for select pollutants (metric tons) for 2017 and 2016 and their respective contributions (%), from the Canadian pulp (NPRI) (Environment and Climate Change Canada 2018) and US paper (AAEI) (Air Emissions Inventory Data 2019) facilities. Data collected separately due to existence of the international border.

| Inventory | Pb | CO | $NH_3$ | $NO_2$ equiv. | $PM_{10}$ | $PM_{2.5}$ | $SO_2$ | VOC |
|---|---|---|---|---|---|---|---|---|
| NPRI 2017 (Canada) | 0.023 | 1774 (99.80) | 75 (99.96) | 993 (99.34) | 79 (96.07) | 56 (97.24) | 1259 (99.71) | 471 (98.23) |
| AAEI 2017 (US) | 5.22E-5 | 2.97 (0.20) | 0.027 (0.04) | 6.55 (0.66) | 3.23 (3.93) | 1.59 (2.76) | 3.64 (0.29) | 8.48 (1.77) |
| Combined 2017 | 0.023 | 1776.97 | 75.027 | 999.55 | 82.23 | 57.59 | 1262.64 | 479.48 |
| NPRI 2016 (Canada) | 0.023 (95.83) | 1768 (99.68) | 70 (99.90) | 1065 (98.75) | 88 (99.78) | 57 (99.70) | 1101 (99.43) | 451 (95.96) |
| AAEI 2016 (US) | 1.35E-3 (4.17) | 5.65 (0.32) | 0.071 (0.10) | 13.48 (1.25) | 0.196 (0.22) | 0.17 (0.30) | 6.31 (0.57) | 19.0 (4.04) |
| Combined 2016 | 0.024 | 1773.65 | 70.071 | 1078.48 | 88.196 | 57.17 | 1107.31 | 470.0 |

*Note:* Percent (%) contributions in parentheses.

**Table 3**
Comparing emissions limits at the Edmundston and Atholville pulp mills in NB, and the Madawaska paper mill.

| Edmundston | | | Atholville | Madawaska | |
|---|---|---|---|---|---|
| Approval I-10081 Emission Limits (Government of New Brunswick, 2018) | | | Approval I-8787 Emission Limits (Government of New Brunswick, 2015) | Total Licensed Annual Emissions (Department of Environmental Protection, 2018) | |
| Substance | Source | Limit | Limit | Source | Limit |
| $SO_2$ | Total | 2000 tonnes/year | 1000 tonnes/year | Total $SO_2$ | 140.5 tonnes/year |
| $SO_2$ | Recovery boiler | 500 ppmv[a] | 500 ppmv[a] | | |
| $SO_2$ | Recovery boiler[b] | 1500 ppmv[a] | 1500 ppmv[a] | | |
| $SO_2$ | Recovery boiler[c] | 1000 ppmv[a] | | | |
| Total PM | Total boilers | 250 tonnes/year | 150 tonnes/year | Total PM | 44.5 tonnes/year |
| Total PM | Recovery boiler | 100 mg/dscm | 70 mg/m$^3$[d] | Total $PM_{10}$ | 44.5 tonnes/year |
| Total PM | Co-gen boiler | 100 mg/dscm | 130 mg/m$^3$ [d,e] | | |
| $NO_x$ as $NO_2$ | Co-gen boiler[f] | 160 kg/h | | Total $NO_x$ | 94.8 tonnes/year |
| $NO_x$ as $NO_2$ | Co-gen boiler[g] | 140 kg/h | | | |
| $ClO_2$ | $ClO_2$ generator stack | 4.0 kg/h | 4.0 kg/h | | |
| $Cl_2$ | $ClO_2$ generator stack | 4.0 kg/h | 4.0 kg/h | | |
| | | | | Total CO | 16.9 tonnes/year |
| | | | | Total VOC | 120.6 tonnes/year |

[a] for any 1 h average;
[b] for power failure events;
[c] if a burndown is necessary to improve scrubber efficiency;
[d] dry gas at 21 degrees C and 101.3 kPa
[e] corrected to 12% $CO_2$
[f] when firing bark or combination fuels
[g] when firing oil only.

in the US required the US EPA to set out air quality standards for "criteria" pollutants (United States Environmental Protection Agency, 2016). As such, CO, lead, $NO_2$, ozone, particle pollution, and $SO_2$ all have maximum allowed concentrations with regulations on how often the levels may be exceeded over various averaging time periods (United States Environmental Protection Agency, 2016). Table 4 compares contaminant thresholds between US and NB regulations, which are generally similar but differ significantly for $SO_2$ and hourly $NO_2$ and have non-identical chemicals of concern.

$SO_2$ emissions from various parts of the facility must meet air quality emissions standards as stated in the Approval to Operate (Government of New Brunswick, 2018). While annual emissions generally meet the Approval standard, 1 h emissions limits for $SO_2$ tend to exceed the limit of 500 ppm at least once per year (New Brunswick Department of Environment and Local Government, 2018). Further, ambient $SO_2$ concentrations at the Cormier School monitoring site have an average number of exceedances that is greater than one per year between 2007–2017 (New Brunswick Department of Environment and Local Government, 2018). It seems likely that an examination of $SO_2$ levels in Madawaska when winds are toward the border would be over the US EPA limit even with the dilution of travelling from the Edmundston facility relatively often, given the disparity of the 1-hour limits, but we currently have no way of investigating this as only PM measurements could be found for Madawaska.

Other contaminants are also being monitored and some exceedances of these are reported as well. For instance, $ClO_2$ and chlorine gas emissions have tended to exceed the 4 kg/h limit at least once per year between 2009–2016 (New Brunswick Department of Environment and Local Government, 2018). However, some contaminants are reported with no apparent examination of exceedances, including some reports to the US TRI (United States Environmental Protection Agency, 2019) and AAEI (Air Emissions Inventory Data, 2019). As most contaminant limits are for time periods shorter than one year, these databases are difficult to compare to exceedance limits.

## 4. Current management and mitigation strategies

Several programs to manage and mitigate various pollutants have been in play in the last few decades. $SO_2$ has been viewed as a problem contaminant due to its role in acid rain production for decades,

**Table 4**
Comparing the US *Clean Air Act* with the NB *Clean Air Act* air quality standards for specified contaminants.

| | US *Clean Air Act* | | NB *Clean Air Act* | | |
|---|---|---|---|---|---|
| | Averaging period | Limit[a] | Averaging period | Limit | Converted Limit[b] |
| **CO** | 1 h | 35 ppm | 1 h | 35000 μg/m$^3$ | 30 ppm |
| | 8 h | 9 ppm | 8 h | 15000 μg/m$^3$ | 13 ppm |
| **$H_2S$** | Not on list | | 1 h | 15 μg/m$^3$ | 11 ppb |
| | Not on list | | 24 h | 5 μg/m$^3$ | 4 ppb |
| **$NO_2$** | 1 h | 100 ppb | 1 h | 400 μg/m$^3$ | 210 ppb |
| | | | 24 h | 200 μg/m$^3$ | 105 ppb |
| | 1 year | 53 ppb | 1 year | 100 μg/m$^3$ | 52 ppb |
| **$SO_2$** | 1 h | 75 ppb | 1 h | 900 μg/m$^3$ | 339 ppb |
| | 3 h | 0.5 ppb | | | |
| | | | 24 h | 300 μg/m$^3$ | 113 ppb |
| | | | 1 year | 60 μg/m$^3$ | 23 ppb |
| **Total suspended particulate** | Not on list | | 24 h | 120 μg/m$^3$ | |
| | Not on list | | 1 year | 70 μg/m$^3$[c] | |
| **$PM_{10}$** | 24 h | 150 μg/m$^3$ | Not on list | | |
| **$PM_{2.5}$** | 1 year | 12 μg/m$^3$ | Not on list | | |
| **Ozone** | 8 h | 0.070 ppm | Not on list | | |
| **Pb** | 3 months (rolling) | 0.15 μg/m$^3$ | Not on list | | |

[a] These limits are more specific with regards to primary/secondary standards and the frequency of possible exceedances
[b] These have been rounded to the nearest integer
[c] This is a geometric mean.

and the Process Point Source $SO_2$ Reduction Program, which ran from 1993 to 1995, was one management plan used to address part of this problem (New Brunswick Department of Environment and Local Government, 2018). Since then, emission caps for $SO_2$ and PM have been introduced (Government of New Brunswick, 2018; New Brunswick Department of Environment and Local Government, 2018). According to the NB facility profile, annual $SO_2$ and PM emissions were below the cap for each year between 2007–2016 (later years were not available for the report) (New Brunswick Department of Environment and Local Government, 2018). The current Title *V* Permit limits the maximum sulphur content in the fuel oils available for burning to 0.5% by weight as of 2018, which reduces sulphur and hence possible related emissions (Department of Environmental Protection, 2018).

Dioxins and furans are pollutants particularly hazardous to ecological and human health (Hoffman et al., 2019). One way to reduce the amount emitted is to switch from elemental chlorine to $ClO_2$ as a bleaching agent in the pulping process (Cheremisinoff and Rosenfeld, 2010). The pulp mill made this switch in 2016 (New Brunswick Department of Environment and Local Government, 2018). This is much later than was required by the *Pulp and Paper Effluent Regulations* (PPER, 2021). Another current strategy to reduce emissions is to ban or restrict the use of certain machinery that does not comply with current standards. The Title *V* Permit currently disallows one boiler from operation entirely and restricts another to operate at a fraction of its rated annual capacity (New Brunswick Department of Environment and Local Government, 2018; Department of Environmental Protection, 2018).

The current Approval to Operate required Twin Rivers Paper Company to develop an action plan to meet the new standards for $SO_2$, $NO_2$, and $PM_{2.5}$ from the Canadian Ambient Air Quality Standards (CAAQS) that came into effect in 2020 (Government of New Brunswick, 2018). Unfortunately, this action plan is not currently publicly accessible.

## 5. Recommendations

As we have mentioned several times throughout this paper, there is a lack of independently verified monitoring of air pollutants. We recommend that more local air quality stations be set up and maintained by people not affiliated with the mills. Further, these stations should monitor VOCs, $SO_2$, and other compounds that are put out by the mill, and not only focus on fine PM as many existing stations do. These could be spaced out to gain a better understanding of the regional effects of the air pollution from the mill, particularly considering the predominant wind directions, as suggested for another mill by Hoffman et al. (2017).

We also recommend that studies be carried out measuring the health impacts on the community due to the air pollution released by the mill. It would be interesting to find how far out effects are felt, and whether their magnitude changes with exposure to various pollutants. Finally, we recommend that studies be carried out on local fauna to examine whether air pollution from the mill has any effect on these, as this is generally missing from the discussion of air pollution in the literature. Lichen chemistry or diversity studies could be used to help examine magnitude and extent of air pollution impacts (Walker et al., 2006; Walker et al., 2009; Walker et al., 2003; Das et al., 2013).

Recommendations to improve air quality need to consider costs to implement changes. It is often possible to add scrubbers or precipitators to stacks to help control particulate emissions (Cheremisinoff and Rosenfeld, 2010). Scrubbers remove gases and particulates from exhaust steam by using a drop in pressure to liquify the gases and expecting solid particulates to be entrained in the liquid; steam can then escape via the stack. Precipitators use an electrostatic charge to remove solid particles from exiting air (Cheremisinoff and Rosenfeld, 2010). This has already been done to some extent but could probably be furthered (Department of Environmental Protection, 2018). In some cases, it may make sense to get rid of equipment that is no longer allowed to operate (*e.g.*, boiler #7) or which can only be operated less than half of the time (*e.g.*, boiler #6) due to the large amounts of pollutants they create (New Brunswick Department of Environment and Local Government, 2018; Department of Environmental Protection, 2018).

Other mills have previously instituted a black liquor chemical recovery furnace which reduces sulphur emissions and produces gains in energy efficiency (Cheremisinoff and Rosenfeld, 2010). This could be an expensive but potentially worthwhile investment, particularly as it reduces odorous compounds, which local communities are frequently concerned about (Jaakkola et al., 1999). Several newer regulations instituted by Title V refer to the maximum sulphur content in fuel oil by weight–further reducing the sulphur content of purchased oil would further contribute to lowering sulphur emissions (Department of Environmental Protection, 2018).

There are many possible ways to decrease the air pollution coming from pulp and paper mills, but many of these are cost-prohibitive or

the costs are generally regarded as outweighing the improvements in air emissions. For instance, the pulp mill could switch to being totally chlorine-free rather than simply free of elemental chlorine, and further reduce its production of dioxins. Additionally, sulphite pulping could be reduced in favor of increasing the pre-existing mechanical pulping process to reduce air pollution (Cheremisinoff and Rosenfeld, 2010).

## 6. Limitations

The major limitation to many of the possible improvements of air quality due to actions taken by the mills is expense. Switching the pulp mill to being totally chlorine-free would be expensive to implement and could reduce profits due to the pulp responding to different processes in different ways, limiting the types of paper that could be produced. Similarly, any reduction in sulphite pulping in favor of the mechanical pulping process which is already on-site would again affect the pulp produced (Cheremisinoff and Rosenfeld, 2010). One major limitation to reducing air pollutants produced by the pulp and paper mills is the age of many of the machines, some of which were installed as far back as the 1920s (Department of Environmental Protection, 2018). Many of these machines have more modern equivalents that are better designed for reducing particulate emissions, but replacing the machines and retrofitting is expensive (Cheremisinoff and Rosenfeld, 2010; Department of Environmental Protection, 2018).

## 7. Conclusions

Pulp and paper is an important industry in NB and ME, but creates many environmental challenges. Unfortunately, it produces many contaminants that are released as air pollution. Facilities often exceed the regulated emissions rates, and sometimes appear to not suffer any consequences for this. Further, the mills appear to be quite slow at adapting to updating legislation, as in the case of the PPER (1992) regulations (PER, 1992). This has put and is currently putting the health of workers and community members at risk, though the extent of the risk is currently badly quantified due to the lack of local epidemiological studies and the lack of air pollution data that is not self-reported by the facilities in question. While expense is a limitation, we have made recommendations as to how this situation could be improved in the future.

## Declarations of Interest

None.

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## Availability of data and material

All datasets used are publicly available.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envc.2021.100245.

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# Ambient hydrogen sulfide, total reduced sulfur, and hospital visits for respiratory diseases in northeast Nebraska, 1998–2000

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This analysis examined associations between total reduced sulfur (TRS) and hydrogen sulfide ($H_2S$) levels, and hospital visits for respiratory disease among residents of Dakota City and South Sioux City, Nebraska, from January 1998 to May 2000. For reference, the association between TRS, $H_2S$, and digestive diseases was also examined. Time-series analyses of daily hospital visits in the selected outcome categories and measures of TRS and $H_2S$ were performed using generalized additive models with a Poisson link. TRS and $H_2S$ levels were categorized as *high* if at least one of the daily 30-min rolling averages was $\geq 30$ ppb and as *low* if every rolling average was $< 30$ ppb. Loess smoothers allowed for flexible modeling of the time effect and the effect of temperature and relative humidity. The measure of association used was the mean percent change in the average number of hospital visits recorded following a day with a high exposure *versus a* day with a low exposure. For children less than 18 years of age, a positive association was found between asthma hospital visits and 1-day lagged TRS levels. For adults, a positive association was found between asthma hospital visits and $H_2S$ levels on the previous day. A positive association also was found between hospital visits for all respiratory diseases, and $H_2S$ and TRS levels on the previous day for children but not for adults. No association was found between contaminant levels and hospital visits for all digestive diseases. These findings suggest that TRS or $H_2S$ levels may be associated with exacerbations of asthma or other respiratory diseases among the residents of Dakota City and South Sioux City.
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**Keywords:** *asthma, respiratory tract diseases, digestive system diseases, air pollution, hydrogen sulfide.*

## Introduction

The Dakota City and South Sioux City communities are located in northeast Nebraska. These communities are bordered on the north and east by the Missouri River and Sioux City, Iowa. The Nebraska Department of Environmental Quality has identified 13 point sources of total reduced sulfur compounds (TRS) in the Dakota City and South Sioux City area. TRS refer to the combined concentration of sulfur in air from hydrogen sulfide, methyl mercaptan, dimethyl sulfide, and dimethyl disulfide, but Federal and state ambient air monitoring data indicate that hydrogen sulfide ($H_2S$) is the primary component of TRS in the area. Sources of TRS include a large beef slaughter and leather tanning facility with a 582,000 square foot liquid waste treatment complex, a wastewater lagoon for a truck wash, and a small municipal wastewater treatment facilities. At the time of the study, the large liquid waste treatment complex (the largest source) was estimated to release approximately 1900 pounds of $H_2S$ per day (Reyher D., personal communication, United States Department of Justice, unpublished data). Approximately 1000 residents live within one mile of this source. Based on historical sampling, the communities were not exposed to ambient sulfur dioxide ($SO_2$) air pollution at levels of concern. As a result, monitoring for $SO_2$ was discontinued years ago. The local communities have voiced concerns for many years that their health might be adversely impacted by the ''rotten egg'' odor and sulfur air pollution. Area residents reported a variety of health complaints, including increased respiratory and neurologic symptoms. Residents have reported that symptoms sometimes occur when they are awakened at night by a strong odor.

In response to these concerns, the Agency for Toxic Substances and Disease Registry (ATSDR) and the US

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Environmental Protection Agency (EPA) conducted residential air monitoring in August 1996 to determine indoor and outdoor levels of $H_2S$ (ATSDR, 1997). Reliable data from five of six homes sampled showed both elevated ambient and indoor levels of $H_2S$. In Dakota City, repeated peak indoor levels of $H_2S$ were measured at greater than 90 ppb in one home and between 40 and 90 ppb in two others. Generally, the amount of $H_2S$ in the air of American cities is between 0.11 and 0.33 ppb (ATSDR, 1997). All five homes had at least 5 ppb of $H_2S$ for a minimum of 75 min during a 2-month period. The findings suggested a correlation between indoor $H_2S$ levels and ambient $H_2S$ levels. However, the available air monitoring data were not sufficient to characterize the spatial and temporal distribution of $H_2S$ levels in the Dakota City.

In 1998, the Nebraska Department of Environment Quality established a TRS standard of 100 ppb for a maximum 30-min rolling average concentration. There are no international standards for $H_2S$. However, in order to avoid substantial complaints about odor annoyance, the World Health Organization (WHO) recommends that ambient $H_2S$ levels not exceed 5 ppb during a 30-min period (WHO, 1981). In the United States, $H_2S$ was established as a regulated toxic substance and a hazardous substance under the Federal Water Pollution Control Act. A population may be exposed to high levels of $H_2S$ through deliberate release of emissions from pulp and paper mills, natural gas drilling and refining operations, and through hot springs in areas with high geothermal activity.

Recent publications that reviewed the toxicity of $H_2S$ are ATSDR (1999) and Milby and Baselt (1999a, b). Many studies have addressed the potential long-term health effects of $H_2S$ and TRS. In studies of subjects exposed to $H_2S$ or TRS, researchers have identified respiratory symptoms, decreased pulmonary function test results, and increases in pulmonary disease. The groups studied were sewer workers (Richardson, 1995), oil and gas workers (Hessel et al., 1997; Mostaghni et al., 2000), and general populations (Dales et al., 1989; Jaakkola et al., 1990; Partti-Pellinen et al., 1996; Legator et al., 2001).

The potential health impact of lower concentrations of $H_2S$ are less well understood. Short-term symptomatic effects have been observed in small experimental studies of healthy volunteers exposed to $H_2S$ (Jappinen et al., 1990; Bhambhani et al., 1997) and in communities exposed to malodorous sulfur compounds (Haahtela et al., 1992; Marttila et al., 1995; Jaakkola et al., 1999). Haahtela et al. (1992) analyzed responses to a questionnaire given to 75 residents after a high $H_2S$ exposure period (maximum 4-h of 135 $\mu g/m^3$; 96 ppb) and after a low exposure period. The occurrence of self-reported ocular, respiratory, and neuropsychological symptoms was considerably increased following the high exposure period. Marttila et al. (1995) found that daily TRS levels (daily average ranging from 0 to 82 $\mu g/m^3$; 0 to 58 ppb) were associated with increases in symptoms of nasal and pharyngeal irritation in a small population living near a pulp mill (Marttila et al., 1995). Jaakkola et al. (1999) reported that a reduction in the emission of malodorous sulfur compounds was related to a significant decrease in nasal symptoms and cough frequency (Jaakkola et al., 1999).

Finally, a significant increase in respiratory disease mortality was observed among Rotorua Maori females living in a geothermal area for the years 1981–1990 when compared to other females in New Zealand (Bates et al., 1997). However, the authors concluded that no convincing evidence of elevated rates of mortality was found because of the possibility of confounding by ethnicity. When Bates et al. (1998) compared the hospital discharge data of Rotorua residents during this time period to that of all other New Zealand residents, they found a significant increase in circulatory diseases among the Rotorua residents (Bates et al., 1998). A recent study from the same authors in the same study area suggests the existence of chronic health effects from $H_2S$ exposure (Bates et al., 2002).

From February 1999 through May 2000, ATSDR and EPA conducted extensive air monitoring to characterize geographic and temporal variation in $H_2S$ levels throughout the Dakota City and South Sioux City area (Inserra et al., 2002). This monitoring was designed to support a neurobehavioral study, but the availability of extensive $H_2S$ data presented a unique opportunity to explore the association between ambient $H_2S$ levels and hospital visits for respiratory illnesses.

The study area included Dakota City and South Sioux City and the study time period was from January 1998 to May 2000. The purpose of the study was to evaluate the relationships (by age group) between malodorous sulfur compounds and specified respiratory health outcomes. The air contaminants of interest were measured as ambient TRS levels, ambient $H_2S$ levels, and indoor $H_2S$ levels. The specific objectives were to investigate associations between the air contaminants of interest and hospital visits for asthma and for all respiratory diseases. As a comparison, associations were also examined between the air contaminants of interest and hospital visits for all digestive diseases.

## Population and methods

### *Source Population*

Dakota City and South Sioux City have a combined population of approximately 14,000 (2000 US Census; http://tiger.census.gov/). The population for this analysis was restricted to persons who sought emergency or nonscheduled outpatient care from January 1998 to May 2000 at the two local hospitals (located in Sioux City) and who resided in zip codes 68776 or 68731. Approximately 68% of the residents with these zip codes live inside the South

Sioux City and Dakota City limits according to the US Census data (Figure 1).

*Data*

The following data were used: (a) the dates and arrival times of emergency visits and nonscheduled outpatient visits, (b) measurements of air contaminants, and (c) weather information. $H_2S$ and TRS levels were usually higher throughout the evening and night hours (see Figures 2 and 3); therefore, a day unit in this investigation corresponds to 24-h period from noon to noon. Weekend periods begin at noon on Friday and end at noon on Monday. Holiday periods begin at noon the day before and end at noon the day after.

*Hospital Visits*

In this investigation, ''hospital visits'' include emergency room and nonscheduled outpatient visits. Daily counts of hospital visits for respiratory diseases or symptoms (International Classification of Diseases, 9th revision, ICD-9: 460–519.9 or 786–786.9) and digestive system diseases or symptoms (ICD-9: 520–579.9 or 787–787.9) between January 1, 1998 and May 31, 2000 were extracted from the computerized billing systems of the two local hospitals. For this study, a patient was categorized as an asthma case if either the primary or secondary ICD-9 codes were 493–493.9 for ''asthma'', 519.1 that includes ''reactive airway disease'', or 786.09 for ''wheezing'' (Tolbert et al., 2000).

In a survey of patients seen in the outpatient clinic of Hospital A during July 2000, approximately 43% of them were scheduled in advance. Scheduled patients were usually those people having routine outpatient procedures, such as endoscopy. Therefore, visits to the outpatient area of Hospital A were not included in this analysis. At Hospital B, however, patient visits to the outpatient clinic visits are not scheduled; instead, patients in distress come to a common area and are triaged to either emergency care or to outpatient care. Visits to the outpatient area of Hospital B were included in this analysis.

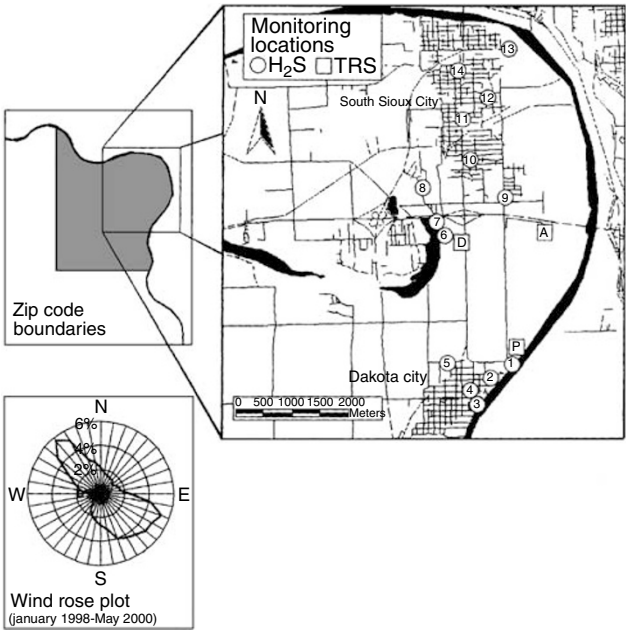


**Figure 1.** Map of the Dakota City and South Sioux City, Nebraska, area indicating the $H_2S$ and TRS monitoring stations and zip codes boundaries. According to the 1996 Bureau of Census estimates, approximately 68% of the inhabitants with these zip codes (68776 and 68731) live inside the South Sioux City and Dakota City limits. Wind rose plot designed from measurements recorded at the Sioux City, Iowa airport (approximately one mile southeast of Dakota City) from January 1998 to May 2000. Prevailing winds are from the south-east during summer time and from the north-west during the rest of the year (data not shown).

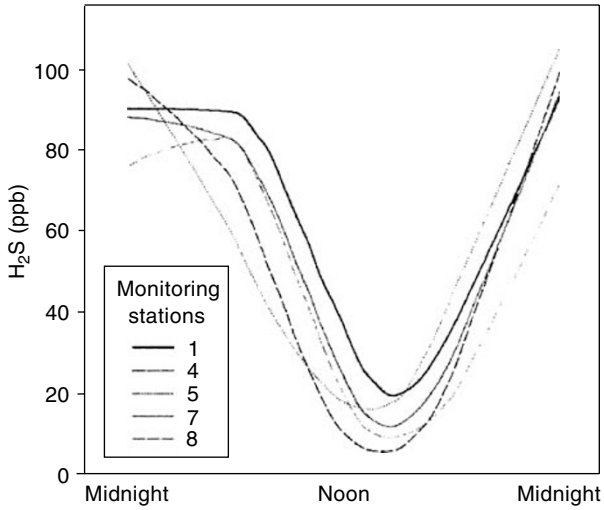


**Figure 2.** Maximum $H_2S$ during the day (loess smoothing of the highest 5 min levels).

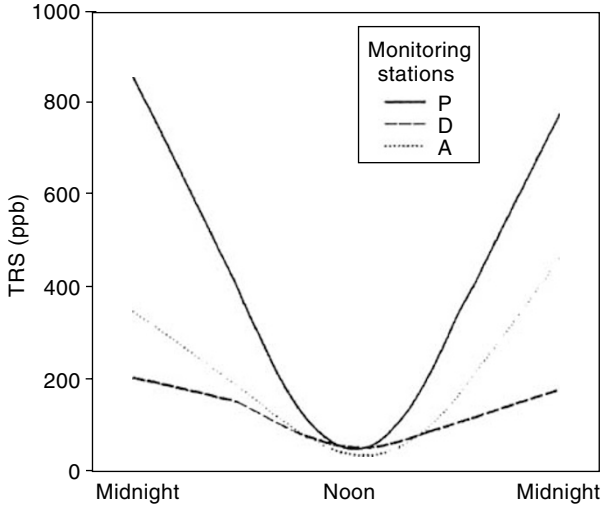


**Figure 3.** Maximum TRS during the day (loess smoothing of the highest 1 min levels).

Visits for digestive system diseases were examined to test for statistically derived associations between health measures and air pollution that are believed to be physiologically implausible.

*Air contaminants*

Figure 1 gives the relative locations of the 19 monitoring stations (three for TRS and 14 for $H_2S$). Because of lack of confidence, results do not include location 6. Instead, this monitor was re-established as ambient monitoring location 7 in July 1999. Ambient TRS levels were measured at three stations every minute between January 1, 1998 and May 31, 2000 by the Nebraska Department of Environment Quality. TRS was sampled using the fluorescent method (EPA EQSA-0495-100 method code API 100 ATRS) (USEPA, 1997).

Ambient and indoor $H_2S$ levels were measured every 5 min between February 5, 1999 and May 31, 2000 by the EPA Environmental Response Team (ERT). A total of 16 Zellweger Single Point Monitors® (SPM) with ChemKey® and Chemcassette® detection system were deployed in eight residences and eight outdoor environmental enclosures. This method is best described as absorbance detection of lead sulfide formed by the reaction of "sulfide" with lead acetate-impregnated tapes. These SPM were configured with $H_2S$ low-level VIP ChemKey® designed by the manufacturer to cover a range of 2–90 ppb. Indoor monitors were placed in first floor living areas in each residence, and operated according to the manufacturer's instructions (Zellweger Analytics, 1995). Sampling locations were selected by ATSDR and EPA, considering: (a) the location of 13 point sources of TRS and their proximity to populated areas, (b) public complaint, (c) accessibility of household, (d) meteorological conditions, and (e) the results of the previous ATSDR exposure investigation (ATSDR, 1997).

Detailed information on characterization of $H_2S$ exposure in Dakota City and South Sioux City was presented elsewhere (White et al., 1999; Inserra et al., 2002). In all, 30-min rolling averages were computed for each TRS and $H_2S$ monitoring station. Levels were characterized as high if at least one of the daily rolling averages was $\geq 30$ ppb, and low if all rolling averages were $<30$ ppb. This cutoff corresponds to the ATSDR minimal risk level (MRL) for an intermediate inhalation exposure (ATSDR, 1999). Dichotomous variables were formed to minimize the effect of missing data. The days with incomplete 24-h monitoring data were handled as follows: days with high exposure (i.e., at least one rolling average was $\geq$ than 30 ppb of TRS or $H_2S$) were included; and days with only low exposures (i.e., all rolling averages were $<30$ ppb of TRS or $H_2S$) were excluded from the analysis.

*Weather*

Temperature, relative humidity and wind direction measurements for the nearest meteorological station (SUX WBAN #14943) located at the Sioux City, Iowa, airport approximately 1 mile southeast of Dakota City, were obtained from the National Climatic Data Center. Of those, less than 1% of temperature and relative humidity measurements were missing and were replaced by data from the next nearest meteorological station, located 60 miles west of Dakota City in Concord, Nebraska. Temperature and relative humidity measurements recorded from the two meteorological stations were highly comparable (data not shown). The daily maximum temperature and maximum relative humidity were included in our analysis.

Figure 1 also gives a wind rose plot designed from hourly measurement (January 1998–May 2000). Prevailing winds are from the south-east during summer time and from the north-west during the rest of the year (data not shown).

*Statistical Methods*

The associations between hospital visits (emergency room and nonscheduled outpatient) and ambient TRS and $H_2S$ levels were analyzed using generalized additive models with a Poisson link because of the non-Gaussian distribution of the data. The measure of association used was the mean percent change (MPC) (Goldberg et al., 2000). The MPC is the percent change in the average number of hospital visits recorded following a day with high exposure *versus a* day with a low exposure. The average number of hospital visits per day was modeled adjusting for time effects, temperature, relative humidity, weekends, and holidays. Loess smoothers were used to allow for flexible modeling of the time trends as well as the effect of temperature and relative humidity. The usefulness of generalized additive models for time-series data using loess smoothers has been previously demonstrated (Schwartz, 1996, 1999; Goldberg et al., 2000). In our analysis, 0-, 1-, and 2-day lags were considered and the one that yields a smaller residual deviance (and thus a better fitting model) was selected. The span required for using the loess smoother was chosen based on minimizing Akaike's information criterion (AIC) (Schwartz, 1996). For time effects, the autocorrelation of the residuals was also considered. A constant dispersion parameter was used to adjust for possible over or under dispersion. The MPC and dispersion parameter were estimated by maximizing the quasi-likelihood. The MPC is computed as $[\exp(\beta)\text{-}1]100\%$. Confidence intervals were constructed taking the over or under dispersion into account by assuming that the estimated regression coefficient was normally distributed. The estimates were obtained using the "gam" function in S-Plus v. 6.1 (Insightful Corporation, 2002). The convergence parameters for the "gam" function were chosen based upon the recommendation of Dominici et al. (2002).

Smoothing involves some form of local averaging of data. The most commonly used techniques involve moving average smoothing, which replaces each element of the series by either the simple or weighted average of $n$ surrounding elements,

where *n* is the width of the smoothing "window". Generalized additive models allow for some variables to be fit through smoothing. This allows for nonparametric adjustments for nonlinear confounding effects such as time trends, season effects, and weather-related variables. Commonly used methods of smoothing include smoothing splines and loess smoothers. Loess smoothers are a moving regression smoother, which is a generalization of a weighted moving average. Loess smoothers do not require a prespecified number of knots at known locations, as is the case for smoothing splines, and are thus considered to be a more nonparametric alternative to smoothing splines.

A stagewise modeling strategy consistent with that of Hagen et al. (2000) was used. The time effect was modeled first using a loess smoother. Temperature and relative humidity were also modeled using a loess smoother. An indicator variable was included that took the value of 1 for the time period from noon Friday and ending at noon Monday; otherwise, this variable took a value of 0. Another indicator variable was included that took the value 1 for various holidays (Christmas, Easter, Thanksgiving, Labor Day, Memorial Day, and the Fourth of July) and 0 otherwise. After taking the background variation of hospital visits into account, the pollution measures for the various stations were introduced. Models were considered for one station at a time. The stations were not combined because of missing and uncorrelated values, and because noncombined measurements reflect exposure levels and exposure variations to the people living nearby.

**Table 1.** Days with high (30-min rolling average $\geq 30$ ppb) and low ($< 30$ ppb) ambient $H_2S$ or TRS levels.

| Monitoring station | | Number of days[a] High exposure | Low exposure | Invalid data |
|---|---|---|---|---|
| TRS (882 monitoring days) | | | | |
| | P | 155 | 450 | 277 |
| | D | 95 | 400 | 387 |
| | A | 104 | 334 | 444 |
| $H_2S$ (488 monitoring days)[b] | | | | |
| | 1[c] | 43 | 424 | 21 |
| | 2[c] | 9 | 455 | 24 |
| | 3[c] | 1 | 477 | 10 |
| | 4 | 28 | 444 | 16 |
| | 5 | 32 | 401 | 55 |
| | 6 | — | — | — |
| | 7[d] | 55 | 250 | 61 |
| | 7[c] | 18 | 450 | 20 |
| | 8 | 31 | 431 | 26 |
| | 8[c] | 1 | 473 | 14 |
| | 9[c] | 16 | 384 | 88 |
| | 10 | 25 | 448 | 15 |
| | 10[c] | 7 | 430 | 51 |
| | 11 | 8 | 448 | 32 |
| | 12[c] | 0 | 478 | 10 |
| | 13 | 2 | 474 | 12 |
| | 14 | 2 | 408 | 78 |

[a]High exposure day: with at least one 30-min rolling average $\geq 30$ ppb; low exposure day: with every 30-min rolling average $< 30$ ppb; invalid data: days with less than 24 h of monitoring.
[b]$\geq 90$ ppb = upper ceiling limit of detection for $H_2S$ monitor.
[c]Indoor monitor.
[d]Brought online in July 1999.
— Data not reliable.

## Results

### *Ambient Levels by Station*

A high TRS level (30-min rolling average $\geq 30$ ppb) was found on about one-fourth of the valid days at each of the three monitoring stations over the 29-month study period (Table 1). A high TRS level occurred most often at station P and least often at station D. A high $H_2S$ level (30-min rolling average $\geq 30$ ppb) was most often found at indoor station 1 and outdoor stations 4, 5, 7, and 8. Depending on the TRS monitoring station, 31–50% of the days were considered invalid while only 2–18% of days were considered invalid for $H_2S$.

Further analyses were conducted for all TRS stations (P, D, and A); $H_2S$ indoor station 1; and outdoor stations 4, 5, 7, and 8, because high levels were measured during more than 25 days at these stations.

### *Hospital Visits*

The visits to the hospital (emergency room and nonscheduled outpatient clinic) during the study period totaled 455 for asthma, 5009 for all respiratory diseases including asthma, and 2271 for all digestive diseases (Table 2). Children ($< 18$ years) accounted for more than a third of the asthma and respiratory disease visits, and less than a fourth of visits for digestive diseases. The percentage of noninsured patients treated for asthma was higher than the percentage of noninsured patients treated for all respiratory diseases or all digestive diseases. Patients for each of the disease groups were mainly from South Sioux City. Hospital A provided more hospital-based care for asthma and all digestive diseases than hospital B.

### *MPC in Hospital Visits*

The spans used for the time effect were 0.3 for children's asthma, 0.4 for the adult asthma models, 0.1 for total respiratory disease, and 0.2 for digestive disease. For temperature and relative humidity, a span of 0.5 was used for all models. The pollution measures were modeled with a 1-day lag.

The MPC in hospital visits for all respiratory diseases including asthma was significantly increased on days following high TRS levels at station D for children (25%; 95% CI:

**Table 2.** Selected subject characteristics by diseases.

| Characteristic | Asthma[a] ($n=455$) $n$ (%) | All respiratory[b] ($n=5009$) $n$ (%) | All digestive[c] ($n=2271$) $n$ (%) |
|---|---|---|---|
| Sex[d] | | | |
| Male | 214 (47.0) | 2353 (47.0) | 957 (42.1) |
| Female | 239 (52.5) | 2634 (52.6) | 1304 (57.4) |
| Insurance status[d] | | | |
| Govt. coverage | 142 (31.3) | 2015 (40.3) | 893 (39.4) |
| Insured | 227 (50.0) | 2392 (47.8) | 1106 (48.8) |
| Not insured | 78 (17.2) | 533 (10.7) | 236 (10.4) |
| Age | | | |
| $<18$ years | 174 (38.2) | 1805 (36.0) | 482 (21.2) |
| $\geq 18$ years | 281 (61.8) | 3204 (64.0) | 1789 (78.8) |
| Hospital | | | |
| A | 260 (57.1) | 2422 (48.4) | 1352 (59.5) |
| B | 195 (42.9) | 2587 (51.6) | 919 (40.5) |
| City of residence | | | |
| Dakota City | 44 (9.7) | 542 (10.8) | 268 (11.8) |
| SouthSioux City | 411 (90.3) | 4466 (89.2) | 2002 (88.2) |
| Weekday | | | |
| Sunday | 53 (11.7) | 672 (13.4) | 243 (10.7) |
| Monday | 73 (16.0) | 732 (14.6) | 366 (16.1) |
| Tuesday | 70 (15.4) | 755 (15.1) | 310 (13.7) |
| Wednesday | 63 (13.8) | 730 (14.6) | 370 (16.3) |
| Thursday | 72 (15.8) | 765 (15.3) | 364 (16.0) |
| Friday | 61 (13.4) | 668 (13.3) | 340 (15.0) |
| Saturday | 63 (13.9) | 687 (13.7) | 278 (12.2) |
| Year of visit | | | |
| 1998 | 204 (44.8) | 2075 (41.4) | 916 (40.3) |
| 1999 | 172 (37.8) | 1926 (38.5) | 912 (40.2) |
| 2000[e] | 79 (17.4) | 1008 (20.1) | 443 (19.5) |

[a]Asthma includes ICD-9 codes where 493–493.9 for ''asthma'', 519.1 that includes ''reactive airway disease'' or 786.09 for ''wheezing''.
[b]Respiratory diseases or symptoms (ICD-9: 460–519.9 or 786–786.9).
[c]Digestive diseases or symptoms (ICD-9: 520–579.9 or 787–787.9).
[d]May not equal 100% because of missing information.
[e]From January 1 to May 31.

**Table 3.** MPC[a] of hospital visits for days with high ambient level (30-min rolling average $\geq 30$ ppb) versus low ($<30$ ppb).

| | | Children | | Adults | |
|---|---|---|---|---|---|
| | | MPC | 95% CI | MPC | 95% CI |
| **All respiratory diseases including asthma** | | | | | |
| *TRS monitoring stations* | P | 2 | −10–17 | 4 | −6–15 |
| | D | 25 | 9–44 | 4 | −7–16 |
| | A | 4 | −11–22 | 4 | −8–17 |
| *$H_2S$ monitoring stations* | | | | | |
| | 1[b] | 40 | 12–76 | 0 | −17–20 |
| | 4 | 19 | −8–54 | 2 | −18–27 |
| | 5 | 6 | −19–40 | −5 | −24–17 |
| | 7[c] | 26 | 5–51 | −4 | −18–12 |
| | 8 | 31 | 4–66 | −18 | −34–2 |
| **All digestive diseases** | | | | | |
| *TRS monitoring stations* | | | | | |
| | P | 15 | −12–44 | −1 | −13–12 |
| | D | 7 | −21–45 | 13 | −3–31 |
| | A | 22 | −9–62 | −8 | −22–9 |
| *$H_2S$ monitoring stations* | | | | | |
| | 1[b] | −12 | −46–44 | −21 | −38–2 |
| | 4 | 35 | −16–117 | 9 | −17–42 |
| | 5 | 17 | −29–94 | −19 | −39–7 |
| | 7[c] | −3 | −37–47 | −5 | −23–18 |
| | 8 | 30 | −18–112 | 8 | −17–39 |

95% CI: 95% confidence interval.
[a]Adjusted for time effects, temperature, relative humidity, weekend, and holidays (Christmas, Easter, Thanksgiving, Labor Day, Memorial Day, and the Fourth of July).
[b]Indoor monitor.
[c]Brought online in July 1999.

9–44; $P=0.002$). The MPC in hospital visits for all respiratory diseases including asthma was significantly elevated on days following high $H_2S$ levels at indoor station 1 for children only (40%; 95% CI: 12–76; $P=0.002$) and outdoor stations 7 (26%; 95%CI: 5–51; $P=0.015$), and 8 (31%; 95%CI: 4–66; $P=0.020$) (Table 3).

The MPC in hospital visits for asthma alone was significantly increased on days following high TRS level at station D for children (65%; 95% CI: 3–163; $P=0.036$) (data not shown). The MPC in hospital visits for asthma was significantly increased on days following high $H_2S$ levels at outdoor station 4 for adults (83%; 95% CI: 4–221; $P=0.035$), but not for children (data not shown).

The MPC in hospital visits for all digestive disease was not significantly elevated on days following high TRS levels or high $H_2S$ measured at any station for any age group (Table 3).

### *Correlations between TRS and $H_2S$ Levels*

Table 4 presents the median, range, and Spearman correlations from the various station measurements. The values were based on a noon to noon time period. The $H_2S$ and TRS stations located the closest together were positively correlated. For example, TRS station P was positively correlated with $H_2S$ indoor station 1 and outdoor station 4 and 5. TRS stations A and D and $H_2S$ outdoor stations 7 and 8 also were positively correlated.

## Discussion

The small number of hospital visits by the elderly did not allow us to analyze exposure–outcome associations among this sensitive population. However, for study area children less than 18 years of age, the analyses did suggest a positive association between hospital visits for asthma and for all respiratory diseases (including asthma) and TRS levels on the previous day. For children, there was also a positive association between all respiratory diseases (including

**Table 4.** Median, range levels and Spearman correlation coefficients for the 24-h $H_2S$ and TRS levels measured during a period from noon to noon.

| | Median (ppb) | (min–max) (ppb) | *n* | P | D | A | 1[a] | 4 | 5 | 7[b] |
|---|---|---|---|---|---|---|---|---|---|---|
| *IRS monitoring stations* | | | | | | | | | | |
| P | 0.4 | (0–86.4) | 562 | 1.00 | | | | | | |
| D | 1.6 | (0–44.0) | 465 | −0.15[c] | 1.00 | | | | | |
| A | 0.8 | (0–48.8) | 409 | −0.17[c] | 0.35[c] | 1.00 | | | | |
| *$H_2S$ monitoring stations* | | | | | | | | | | |
| 1[a] | 0.0 | (0–31.6) | 467 | 0.59[c] | 0.04 | 0.12 | 1.00 | | | |
| 4 | 0.0 | (0–11.0) | 467 | 0.27[c] | 0.07 | 0.08 | 0.54[c] | 1.00 | | |
| 5 | 0.0 | (0–15.3) | 432 | 0.26[c] | 0.07 | 0.10 | 0.35[c] | 0.61[c] | 1.00 | |
| 7[b] | 0.2 | (0–19.9) | 303 | 0.13 | 0.48[c] | 0.41[c] | 0.11 | 0.18 | 0.25[c] | 1.00 |
| 8 | 0.0 | (0–15.7) | 462 | 0.01 | 0.38[c] | 0.33[c] | 0.06 | 0.21[c] | 0.28[c] | 0.83[c] |

[a]Indoor monitor.
[b]Brought online in July 1999.
[c]*P*-value $<0.01$.

asthma) and $H_2S$ levels on the previous day. Among adults, a positive association was also found between hospital visits for asthma and $H_2S$ levels on the previous day. Children are more likely than adults to suffer from asthma and may be more susceptible to toxicants. This may explain why more positive associations were found for children than for adults.

These findings are consistent with other studies showing short-term respiratory health effects among small groups of humans (Jappinen et al., 1990; Bhambhani et al., 1997) and in communities (Haahtela et al., 1992; Marttila et al., 1995; Jaakkola et al., 1999). In addition, no association was observed between TRS or $H_2S$ levels and hospital visits for digestive diseases. As in a number of previous studies of air pollution and respiratory illnesses, visits for digestive diseases were used as a comparison diagnostic category.

This study examined the association between TRS and $H_2S$ levels and hospital visits for respiratory diseases in a general population. A strength of this study was the ability to monitor community-wide TRS and $H_2S$ exposure levels. Another strength was the participation of the two local hospitals. It was assumed that most, if not all, hospital visits for Dakota City and South Sioux City residents were captured. Major confounders, such as weekends, holidays, relative humidity and temperature, were also 'included' in the model to control for climatic, temporal, and other potential confounders. Previous sampling by the State of Nebraska and during the ATSDR Exposure Investigation identified $H_2S$ as the only sulfur contaminant present in sufficient concentrations to cause concern (ATSDR, 1997).

This study was, however, limited in several ways. One of the major limitations was the quality of available exposure data. As this is an ecological study, personal exposures, which can vary due to air conditioning and time spent outdoors, were not measured. Therefore, we assumed that ambient levels of TRS or $H_2S$ were meaningfully related to personal exposure. Days with only low exposures (i.e., $<30$ ppb of TRS or $H_2S$) and incomplete 24-h monitoring were excluded in an effort to reduce bias associated with misclassification of exposure. The number of invalid days varied by station, which may partly explain why significant associations were observed with some stations and not with others. Additionally, the source and proximity of each monitoring station to the community differed, which may explain some variability in the associations observed. Since 32% of the residents with Dakota City and South Sioux City zip codes lived outside city limits, this may have resulted in some misclassification of exposure. We also assumed that the patient was located in the study area during the day before he/she went to the hospital.

In the present study, associations with TRS and $H_2S$ levels were presented separately for every monitoring station with at least 25 days of high air contaminant levels. For several reasons, this analytic methodology was preferred to analyses based on exposure data aggregated from the various monitoring stations. First, there was a high frequency of missing data following automatic monitoring equipment failure or electricity outage. Owing to the logistical difficulties of keeping a maintenance technician on site, there were periods of days or weeks between equipment failure and repair and/or calibration by a technician. Completeness of $H_2S$ monitoring data ranged from 100 to 87%, with a mean of 96% (Inserra et al., 2002). Completeness of TRS data was not as good and ranged from 83 to 64% (data not shown). Second, $H_2S$ and TRS measurements with suspected lack of calibration were treated as invalid. Finally, developing an aggregate exposure variable would require replacement of missing values by seasonal averages or by levels recorded at other monitoring stations. We believe that such replacement of missing data could exacerbate any exposure misclassification issues. For example, the main source was located in the middle of the study area (in the middle of a triangle formed by the three TRS monitoring stations). On a windy day, residents living downwind of the main source may be heavily exposed, while residents in other areas may not be exposed at all. In addition, residents more distant to the sources may

have had only sporadic, very low-level exposures (Inserra et al., 2002).

Misclassification of the outcome variable in studies of hospital visits constitutes another important concern, but it is likely to have occurred nondifferentially and thus have attenuated any association that may exist (Zeger et al., 2000). Another limitation is that some affected residents may not have sought medical treatment, or may have sought treatment in facilities other than the two local hospitals. We had no data for these other visits.

In conclusion, these findings are consistent with the hypothesis that exposure to malodorous sulfur compounds increases the risk of respiratory problems and symptoms. Specifically, the findings suggest that ambient levels of TRS and $H_2S$ may have been associated with exacerbations of asthma or other respiratory diseases among residents of Dakota City and South Sioux City during 1998–2000. Further analyses and replication in other exposed communities are needed to confirm these findings. Such confirmation would point to the need to protect children and adults who live near facilities emitting TRS or $H_2S$.

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