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Real Time Monitoring of Airborne Hydrogen Sulfide Using Pulsed Fluorescence SO_2-H_2S-CS Analyzers

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ABSTRACT

The paper describes results from real time monitoring of Airborne Hydrogen Sulfide (H_2S) in NE-Iceland, i.e. the lake Mývatn area, town of Húsavík and the area of Kelduhverfi. In general, the H_2S concentration is low and below regulatory limits in all the areas. Located close to both natural emissions of H_2S as well as geothermal power plants in Bjarnarflag and Krafla, stations in the Mývatn area generally show higher concentrations of H_2S than observed in Húsavík and Kelduhverfi. Stations in Húsavík and Kelduhverfi are not close to any major sources of H_2S, but the Húsavík station shows signs of H_2S emission from the Theistareykir geothermal power plant. Clear relationship is observed between H_2S concentration and weather conditions. For a given station, the highest H_2S concentrations normally are measured during cold weather (<0°C) and light breeze from certain wind directions. Inversion in the troposphere occurs, when a layer of cold air at the surface is overlain by a layer of warmer air, resulting in accumulation of H_2S near the surface. Periods of H_2S accumulation are rare and short term.

1. INTRODUCTION

Landsvirkjun is owned by the Icelandic state and produces 75% of all electricity used in Iceland. In addition to multiple hydro power plants, the company operates three geothermal power plants, in NE-Iceland. The Bjarnarflag (1969) and Krafla (1977) power plants are in the Mývatn area and the Theistareykir (2017) power plant is about 20 km towards north from the lake. In order to monitor the air quality in local settlements, the company has installed four automatic H_2S and weather monitoring stations. Two of these stations, Reykjahlíð and Vogar, are in the Mývatn area and the other two are closer to the Theistareykir power plant, in town of Húsavík and in Kelduhverfi, respectively. Time series of H_2S concentration in NE-Iceland reach back to 2011. The above-mentioned monitoring stations have been in simultaneous operation since 2015.

The H_2S analyzers used in this project operate on the principle that H_2S can be converted to sulfur dioxide (SO_2). The amount of SO_2 molecules in an air sample is proportional to the energy emitted when SO_2 molecules, excited in ultraviolet light, decay to a lower energy state. A built-in converter is used to convert H_2S and any reduced sulfur species to SO_2. The inferred H_2S reading is the difference in SO_2 between two measurements of ambient air, one without and the other after being shunted through the converter[1]. The type of equipment used in Reykjahlíð and Vogar is Airpointer 2D, but Thermo 450i in Húsavík and Kelduhverfi. The built-in converter uses a molybdenum catalyst to convert H_2S to SO_2. Experiments have shown, that the conversion efficiency of the catalyst may be reduced as the relative humidity of air increases. This would lead to under-estimation of the actual H_2S concentration[2]. Instruments used in Iceland have not been tested for relative humidity interference.

Icelandic regulation on the concentration of hydrogen sulfide in atmospheric air[3] is stricter than for example WHO's "Air quality guidelines for Europe "[4]. Due to technical limitations of the monitoring equipment, such as detection limits and zero noise, it can be hard to demonstrate that air quality is within Icelandic regulatory limits.

Figure 1 shows the location of monitoring stations at Reykjahlíðarskóli and Vogar in the Mývatn area, in town of Húsavík and in Kelduhverfi. The geothermal power plants "Kröflustöð", "Bjarnarflagsstöð" and "Þeistareykjavirkjun" are shown as well. The locations of monitoring stations are indicated with a yellow circle and the locations of geothermal power plants with a light blue/white sign.

Figure 1. Geothermal power plants and H_2S monitoring stations in NE-Iceland.

2. MEASURED CONCENTRATIONS AND REGULATORY LIMITS

Environmental limits for hydrogen sulfide concentration in atmospheric air are defined in the Icelandic regulation on the concentration of hydrogen sulfide in the atmosphere, regulation no. 514/2010[3]. The limits are divided into two categories, health-limits and a "notification-limit". The limits correspond to the concentration of H_2S in µg/m^3 (micrograms pr. cubic meter) at 293 K and 1013 hPa, see table 1.

Table 1. Environmental limits for airborne H_2S in Iceland.

Environmental limit	Description	Value [µg/m^3]	Deviations
Health limit #1	Daily maximum of 24-hour running average	50	May be exceeded 3 times annually
Health limit #2	Annual mean	5	May not be exceed
Notification limit	Three-hour average	150	Must be reported every time

In practice, the logged concentration of H_2S is a 10 minutes average mole fraction of H_2S in parts pr. billion (ppb). The conversion from ppb to µg/m^3 at 293 K and 1013 hPa is based on the ideal gas law, by multiplying the logged value in ppb with 1.42. The 24-hour running average is calculated every 10 minutes and contains the last 144 logged values. The three-hour average contains the last 18 logged values[5].

As mentioned, the monitoring stations measure the concentration of SO_2 and not H_2S. The inferred H_2S reading is the difference in SO_2 between two measurements of ambient air, one without and the other after being shunted through a converter where a catalyst converts H_2S and any reduced sulfur species to SO_2. For low H_2S concentrations, the accuracy in determining the SO_2 can result in both positive and negative values of H_2S. It is generally recognized that uncertainty in the measurement of H_2S is +/- 3 µg/m^3. Notice, that the uncertainty is of the same magnitude as health limit #2.

Figure 2 shows the H_2S concentration at Vogar and Húsavík, the daily maximum of 24-hour running average from 2015 to 2018[6,7]. A green line indicates the corresponding health limit #1. During the 4-year period, the daily value exceeded the health limit twice at Vogar, in January 20 and 21, 2016, but the limit was not exceeded in Húsavík. The daily maximum of 24-hour running average in Reykjahlíð and Kelduhverfi never exceeded the limit.

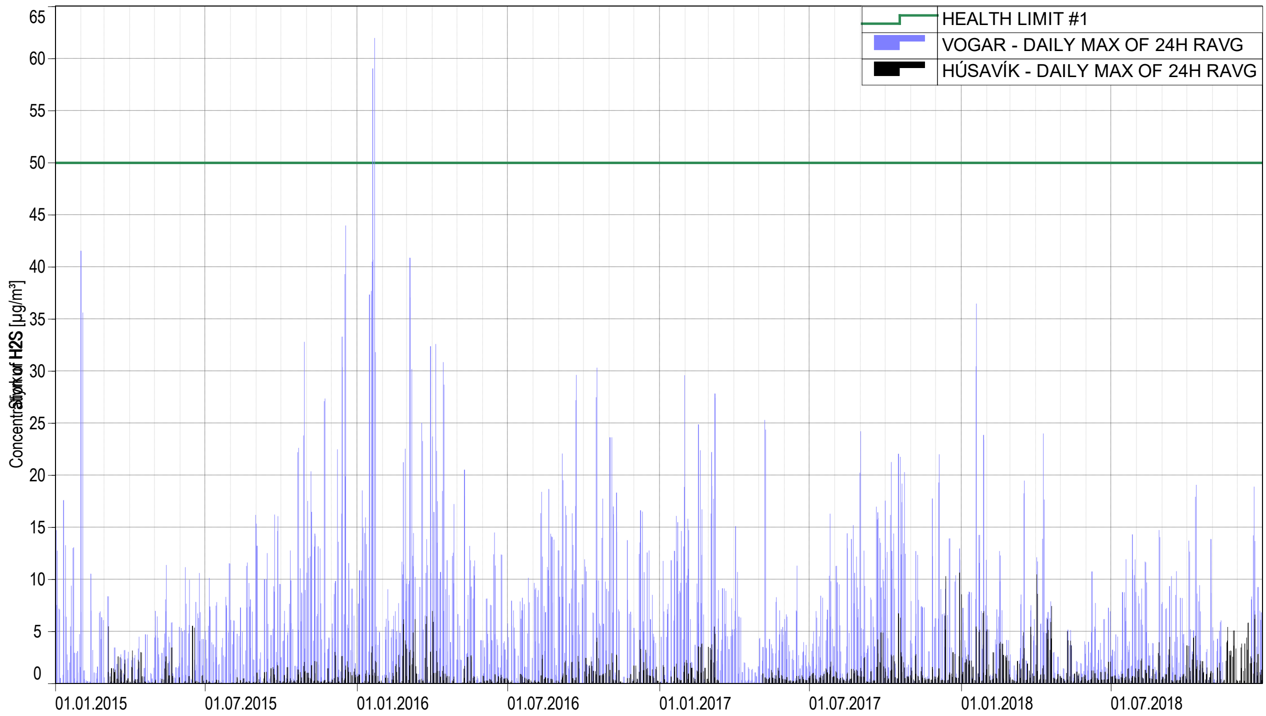

Figure 2. Concentration of H_2S in Vogar and Húsavík, daily maximum of 24-hour running average.

The annual mean values of H_2S concentration are shown in table 2. Compared to the regulatory health limit #2, none of these values exceeded the limit during 2015 to 2018.

Table 2. Annual mean values of H_2S in µg/m3, 2015 through 2018.

YEAR	VOGAR	REYKJAHLÍÐ	KELDUHVERFI	HÚSAVÍK
2015	3	3	1	0
2016	5	5	1	0
2017	4	4	1	1
2018	3	3	1	1

3. THE EFFECT OF GEOGRAPHIC LOCATION AND WEATHER CONDITIONS.

In order to understand the variability in the measured H_2S concentration, it is useful to look further into both the geographic location of the relevant monitoring stations as well as the weather conditions. As a case study, the following subsections will discuss these effects on measurement results from Vogar in 2016.

3.1 Geographic location

The geographic location of the monitoring station in Vogar is shown in figure 1. The station is located next to Mývatn and close to both natural emissions of H_2S as well as geothermal power plants and steam wells emitting H_2S. The Krafla plant is located 11 km towards northeast (36°), the Bjarnarflag plant is 4 km towards east-northeast (60°) and steam wells in Bjarnarflag are at around 70°. A new plant is planned in Bjarnarflag at 80° but has not been built. Figure 3 shows the H_2S concentration in Vogar as a function of wind direction, as measured during 2016. The red line shows the maximum concentration for 90% of measured values within each 5° in wind direction.

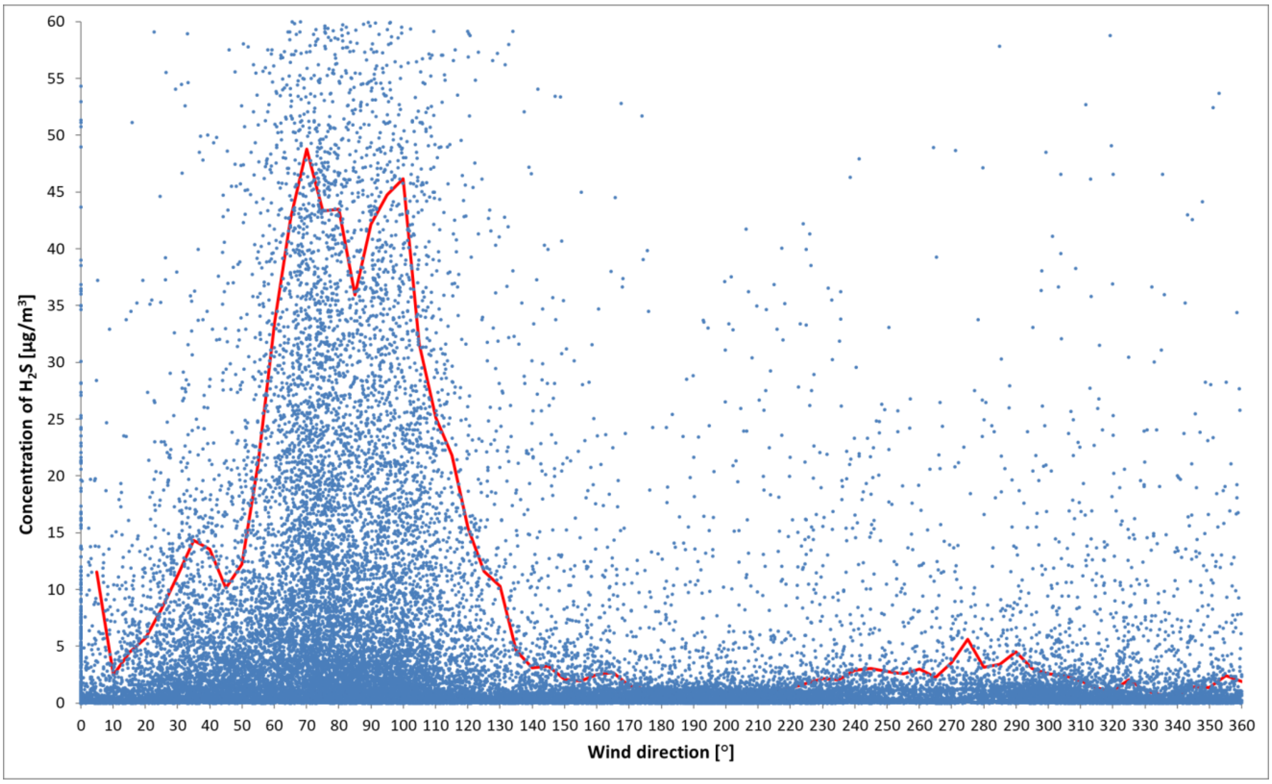

Figure 3. Concentration of H_2S in Vogar as a function of wind direction, 2016.

By comparison of figures 1 and 3 it can be concluded, that the Krafla power plant (36°) most likely has some effect on H_2S concentration in Vogar and the Bjarnarflag area (60-70°) obviously has significant effect. On the other hand, there are no steam wells located further east from Vogar. The response in H_2S concentration at 90 to 100° most likely stems from natural emissions.

3.2. The effect of wind speed and air temperature on H_2S concentration.

Analysis of weather data from Vogar reveals that during January 16 through 19, 2016 there was calm wind and the air temperature was from -10 to -15 °C. These weather conditions may have caused temperature inversion, a layer of cool air at the surface was overlain by a layer of warmer air and the dispersion of H_2S was reduced. The wind increased the following days, blowing from 80 to 100°, moving the accumulated H_2S towards Vogar.

Figure 4 shows the average concentration of H_2S in Vogar as a function of wind speed. The average is calculated for every 1 m/s change in wind speed. It is observed that the concentration of H_2S is reduced with increased wind speed. Given the amount of H_2S released is constant with time, the results show increased dilution and dispersion of emissions, proportional to wind speed.

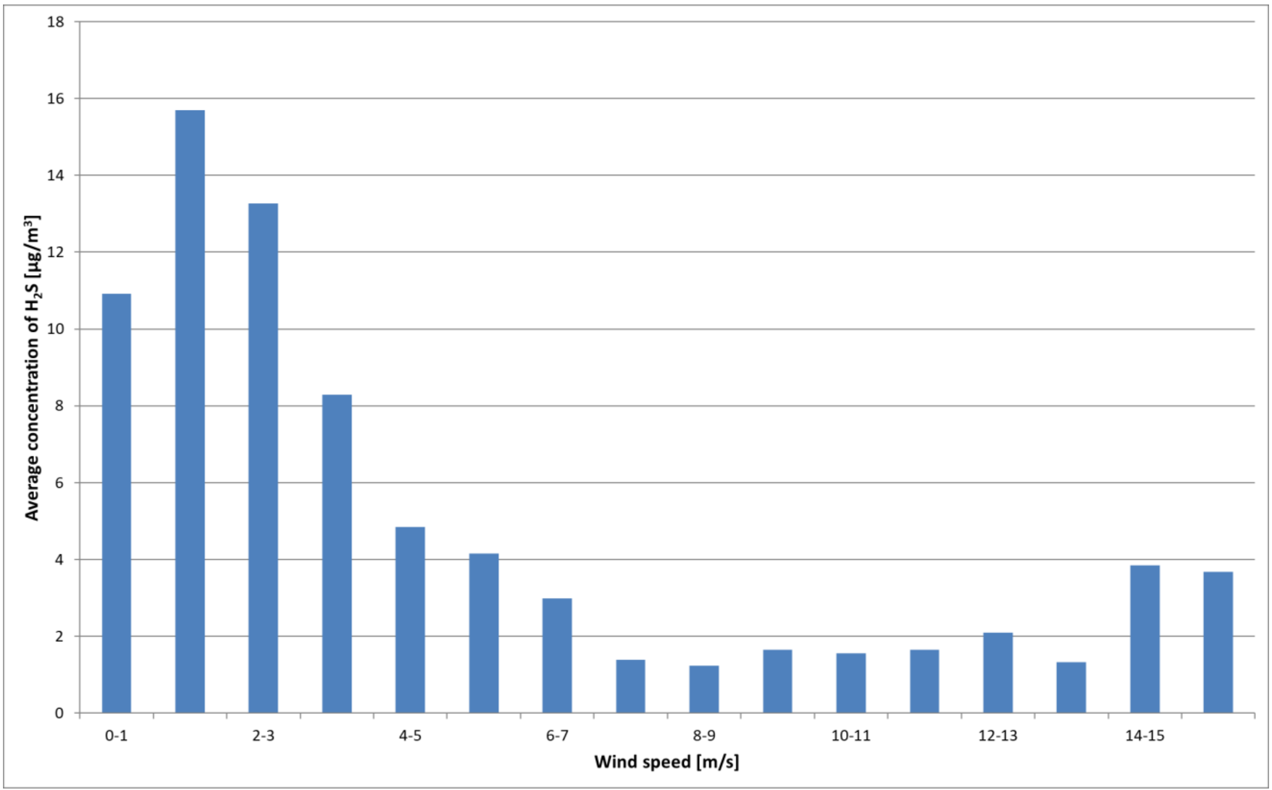

Figure 4. Average concentration of H_2S in Vogar as a function of wind speed, 2016.

Figure 5 shows the average concentration of H_2S in Vogar as a function of air temperature. In general, the concentration of H_2S is inversely related to the temperature of air. Several factors may influence the relationship. Despite wind speed and direction, precipitation above 0°C should affect the concentration, given the high solubility of H_2S in water[8]. Below 0°C the effect of precipitation most likely is milder. Relative humidity (RH) of air might influence the results, as mentioned in the Introduction. The variability in RH is large for most temperatures and therefore the effect of RH on H_2S values should be random. An exception is during extremes in air temperature. Both on cold and warm days, the RH is normally reduced which in a given case will result in raised H_2S values. Finally, low temperatures during calm wind periods may cause a temperature inversion in the troposphere, which will increase the concentration of H_2S in geothermal areas.

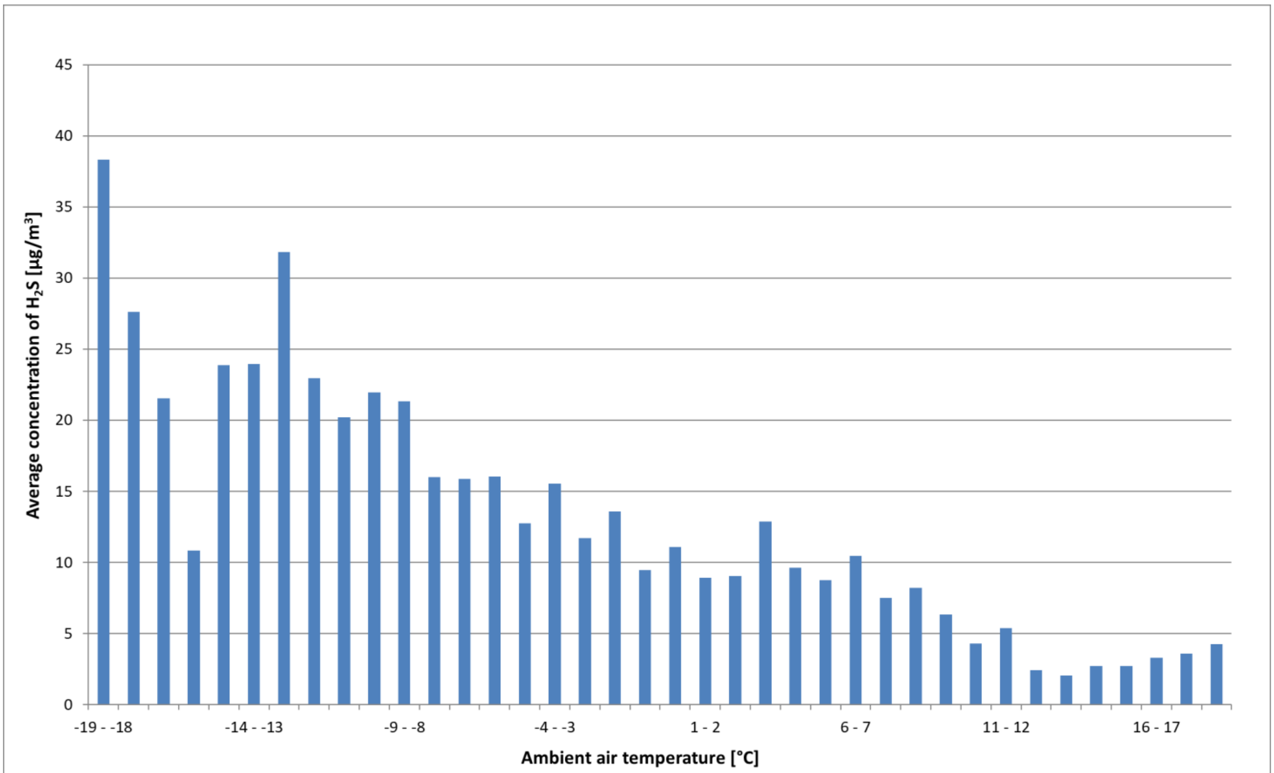

Figure 5. Average concentration of H_2S in Vogar as a function of ambient air temperature, 2016.

4. CONCLUSION

In general, the H_2S concentration in settlements next to utilized geothermal areas in NE-Iceland is low and within environmental limits. Sources of H_2S emissions are both natural and geothermal utilization. When measurement uncertainty of the monitoring equipment is considered, the concentration of hydrogen sulfide in atmospheric air is often on the verge of being measurable. At the same time, the environmental limits specified in the corresponding Icelandic regulation are strict.

Several factors influence the dispersion of H_2S in air. Weather conditions and geographic location of both monitoring stations and sources of H_2S must be considered when evaluating the data. Wind direction is a decisive factor. The highest concentrations of H_2S normally are measured during light breeze at low temperatures.

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