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Near Field Sinks and Distribution of H_2S from Two Geothermal Power Plants in Iceland

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ABSTRACT

Geothermal power production has been expanding in SW-Iceland during the last few decades. The simultaneously increasing emission of hydrogen sulfide (H_2S) has resulted in increased public concern. In this study H_2S was measured in the atmosphere and sulfur in vegetation and precipitation near field of two geothermal power plants, the Nesjavellir and Hellisheidi Power Plants, in southwest Iceland to determine the H_2S distribution and its sinks from the atmosphere. Sinks of atmospheric H_2S, up to 35 km from the Power Plants, were identified and an attempt made to quantify them. Calculations and data analysis indicated that oxidation and uptake in surface water were the largest sinks in the area and washout with precipitation was the smallest sink. Other sinks were uptake by vegetation, uptake by soil and washout in steam at the source. The total estimated depletion of the H_2S from the atmosphere was 256 tons/year which was insignificant compared to the total emission which was over 20,000 tons in 2012. About 99% of the H_2S emissions from the Power Plants were thus transported out of the study area as H_2S. The results from atmospheric measurements of H_2S showed that the H_2S concentration was influenced by weather conditions and terrain. The shape of the H_2S plumes observed at the same location was similar between events, indicating repeated patterns of plume distribution. Plumes were found to follow mountain passes and accumulate alongside a mountain range.

1. INTRODUCTION

Emissions of H_2S are increasing worldwide in relation to increasing geothermal utilization as a part of an effort to decrease reliance on carbon based energy. H_2S gas is also produced as a byproduct in various industries, e.g. kraft pulp mills (Bordado and Gomes, 2003), wastewater treatment plants (Latos et al. 2011) and coal gasification plants (Ko et al., 2006). H_2S is a concern in areas near its sources. It can be harmful to human health in high concentrations but its vigorous smell can become a nuisance at low concentrations, or above 7 $\mu g/m^3$ for a 30-min average (WHO, 2000). Different countries have set environmental guidelines or health limits based on the odor nuisance or possible health impacts. In spite of this an overall estimate of its sinks has not been reported.

Watts (2000) stated that there were few data and large uncertainties in the size of the global mass H_2S budget and assumed that the only sink of H_2S in the atmosphere was a reaction with OH radicals. Research on a local scale have indicated other sinks. Kristmannsdottir et al. (2000), Thorsteinsson et al. (2013) and Olafsdottir et al. (2014b) reported a decrease in H_2S concentration during and following precipitation. However, Susaya et al. (2011) found that airborne H_2S showed positive correlations with relative humidity, dew point and rainfall. It has been proposed that H_2S can be harmful to vegetation (Bartiromo et al., 2012, Tretiach and Ganis, 1999,). Also, uptake by vegetation has been reported (Bussotti et al., 1997). Soil has been reported to have a substantial capacity for sorption of H_2S in laboratory experiments (Smith et al., 1974, Cihacek and Bremner, 1990).

It has been observed that wind conditions and terrain features affect the H_2S distribution (Thorsteinsson et al. 2013, Olafsdottir and Gardarsson, 2013, Olafsdottir et al., 2014b).

Geothermal utilization has been increasing in Iceland during the last few decades and it is still increasing. Two geothermal power plants are within 30 km of Iceland's capital of Reykjavik, the Nesjavellir and Hellisheidi Power Plants, and their production has been expanding with more public concern. In 2010 a health limit was set in Iceland by the Ministry for the Environment and Natural Resources (2010) at 50 $\mu g/m^3$ for a running 24-hour average. This regulation requires the geothermal industry in Iceland to lower H_2S emissions (Gunnarsson, et al., 2013). This paper reports the main findings of a research aimed at estimating the sinks and analyzing the distribution of atmospheric H_2S near field of the two geothermal power plants.

2. STUDY AREA

The study area was within 35 km of the Nesjavellir (180 m a.s.l.) and Hellisheidi (260 m a.s.l.) Geothermal Power Plants in the Hengill volcanic system in Southwest Iceland. The power plants are 10 km apart, located on the north and south side of Mt. Hengill (805 m a.s.l.). Northeast of the mountain is Iceland's largest natural lake, Lake Thingvallavatn (83 km^2). The Nesjavellir Power Plant is located near the lake shore in a small valley, with ridges rising to the west and to the east. The Hellisheidi Power Plant is located at the southwest base of Mt. Hengill with Middalsheidi Heath to the west sloping towards the capital area with the easternmost part about 20 km from Mt. Hengill (Figure 1). Small towns are located within the study area, away from the capital, as well as farms and summer houses. Even so, most of the area is sparsely populated. The local flora is characterized by moss, grass and small shrubs.

The Nesjavellir (NV) Power Plant is about 25 km from the easternmost part of the capital of Reykavik. Hot water production started in 1990 and electricity production in 1998, the current production is 120 MW_e and 300 MW_{th}. The Hellisheidi (HH) Power Plant is about 18 km from the easternmost part of Reykjavik City. Electricity production began in October 2006, the current

installed capacity is 303 MW_e and 133 MW_{th}. The gas is released at the top of the cooling towers of the power plants where the gas is vigorously mixed with the steam in four closely spaced outlets, with fans to accelerate emission. The power plants have expanded the production over the years yielding increased H_2S emissions. The total emission in 2012 was 20,684 tons of H_2S; the yearly emissions from both power plants from 2006 to 2012 is shown in Figure 2. Note that emission from the Hellisheidi Power Plant in 2006 has been calculated from energy production data as a collecting system had not been installed.

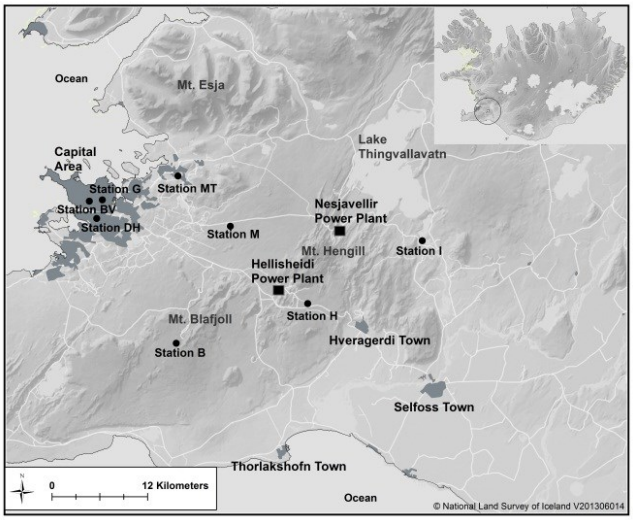

Figure 1: Study area, showing weather stations and other measuring stations used in the study, the capital area and local towns, and the Nesjavellir and Hellisheidi Power Plants. Roads are shown in white; lakes, rivers and ocean are in light gray.

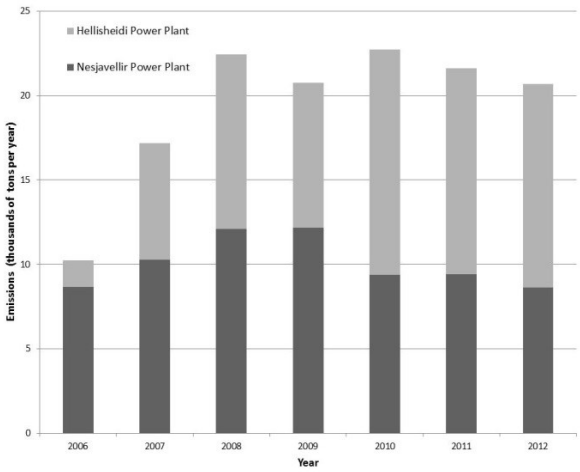

Figure 2: Emissions from Nesjavellir and Hellisheidi Power Plants from 2006-2012 (Thorarinsdottir and Sigurdardottir, 2013). Emission from the Hellisheidi Power Plant in 2006 was calculated from energy production. The 2011 and 2012 emissions were revised by Reykjavik Energy in June 2014.

3. METHODS

3.1 Model

An idealized box model approach was used when estimating the depletion of H_2S from the atmosphere. Spatial variability was neglected and a homogeneous H_2S concentration was assumed in a volume with the horizontal area of a standard Gaussian plume (Figure 3). The source was assumed to be at the center of mass of the combined emissions. The plume's horizontal and vertical dimensions were found by applying a Gaussian plume model in neutral air, over a 35 km distance. The plume was assumed to expand from the source to $4\sigma_y$, 6.57 km, yielding an areal extent of $1.1 \cdot 10^8$ m^2. The plume height was assumed to be represented by $2\sigma_z$ at 35 km or 544 m over the whole area. The mean wind speed at Station H, 7 m/s, was used to find a traveling time out of the area, 83 minutes, during which 3.3 tons of H_2S were discharged into the atmosphere on average in 2012. Assuming homogeneous distribution within the plume volume, this yielded a concentration of C_{air}=55 µg/m^3.

3.2 Measurements

A 12 month measurement program was carried out in 2009 in order to determine the areal extent and strength of geothermal plumes in different weather conditions. H_2S measurements were performed along the major highways (Figure 4) downstream of the

plumes, at different spatial intervals between events. Measurements were made for 3-4 days in a row (randomly chosen) each month for 1-3 hours each day. Events chosen for further analysis all had steady wind direction during the event and good spatial resolution on the road used. The events were measured during different seasons. Measurements were made using a handheld measuring instrument, Jerome 631-X (Arizona Instruments, USA), which has a 4-7100 μg/m^3 detection range and a ±4 μg/m^3 accuracy. Two measurements (with a measurement time of about 20 s) were made at each location and the average concentration was converted using the conversion factor of 1 ppm = 1420 μg/m^3 H_2S (at 20° and 1 atm).

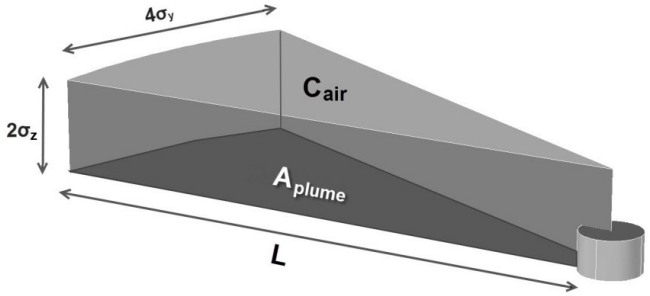

Figure 3: A schematic view of the control volume of the Gaussian plume.

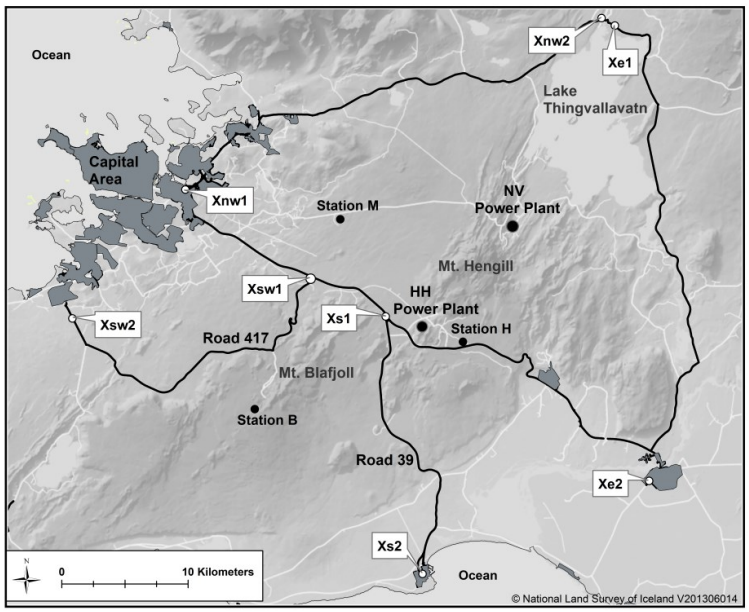

Figure 4. Major highways on which H_2S measurements were conducted.

3.3 Meteorological data

Wind and air temperature data from weather stations at Middalsheidi Heath (Station M), Hellisheidi Heath (Station H), Mt. Blafjoll (Station B) and Mosfellsbaer Town (Station MT), measured at 10 minute intervals, were obtained. Accumulated hourly precipitation was obtained from the weather station at Hellisskard Pass, 2 km northwest of Station H. Ceiling height and cloud cover were measured every 3 hours in Reykjavik, about 30 km from the power plants. Bi-daily upper air data were obtained from Keflavik airport, about 60 km southwest of the power plants. All weather data were provided by the Icelandic Meteorological Office except for data from Station H, owned by the Icelandic Road Administration and from Station MT provided by the Environment Agency of Iceland.

Solar radiation is always moderate or slight at the northerly latitude of Iceland, such that very unstable air conditions rarely occur. In winter, the air can be stable during the day. The Pasquill air stability class during the selected events was estimated by calculating two local air temperature gradients, from ground to the 925 hPa height obtained at Keflavik Airport for both Station M and H, and categorizing them according to Woodward (1999). In addition, the upper air temperature profile at Keflavik Airport at noon was plotted and compared to the adiabatic lapse rate. The three air stability estimates were generally consistent within one stability class.

4. RESULTS AND DISCUSSION

4.1 Sinks of Hydrogen Sulfide

Sinks of hydrogen sulfide identified were: 1) Oxidation; 2) Uptake in surface water; 3) Washout; 4) Uptake by vegetation and 5) Uptake by soil. Estimations of the first three sinks are described below, for more detailed information see Olafsdottir et al. (2014a).

4.1.1 Oxidation

Atmospheric H_2S can be oxidized to SO_2 in a series of reactions starting with the reaction of H_2S with the OH radical.

$$H_2S + OH \rightarrow HS + H_2O \quad (1)$$

In order to estimate the potential oxidation amount of H_2S within the study area the oxidation rate of reaction (1) was used. The general equation for the reaction is

$$\frac{dC_{air}}{dt} = -k_o C_{air} \quad (2)$$

where C_{air} is the H_2S concentration in the atmosphere, $k_o = k_{OH+H_2S} \cdot C_{OH}$, where k_{OH+H_2S} is the rate constant for reaction (1), $5 \cdot 10^{-12}$ cm^3/molecule·sec (Cox and Sheppard, 1980) and C_{OH} is the concentration of the OH radical. The rate of oxidation is thus dependent on the availability of the OH radical. The OH formation is greater during summer as it is dependent on radiation, though here it was assumed to be stable over the year and its concentration constant as the mean of OH concentration found by Spivakovsky et al. (2000) at 60°N latitude over four different months ($2.75 \cdot 10^5$ molecule/cm^3). The OH radicals were assumed to be available only for H_2S oxidation. The oxidation mass rate within the plume is:

$$\frac{dM_{air}}{dt} = k_{OH+H_2S} C_{OH} C_{air} V_{plume} \quad (3)$$

where M_{air} is the mass of H_2S in the plume, V_{plume} is the volume of the plume. These assumptions yield an oxidation of H_2S of 143 tons in one year or 0.69% of the 2012 emissions.

This is a conservative estimate as OH is very reactive and reacts with various other chemicals in the atmosphere and is therefore not necessarily available to react with H_2S. Also, the OH formation is likely to be slower than assumed, especially in the middle of the plume, where the geothermal gases are dense.

The above calculations indicate that H_2S is not oxidized within the study area in significant amounts compared to the 2012 emissions.

4.1.2 Surface water uptake

Wind direction is towards the Lake Thingvallavatn 10% of the time as shown in Table 3. The atmospheric H_2S can be dissolved in the lake when the plume lies over it. The flux of H_2S through the surface of the lake is modeled with

$$\frac{dM_{water}}{dt} = k_w A_{water} dC \quad (4)$$

where M_{water} is the mass of H_2S dissolved in the lake, k_w is the gas transfer velocity, A_{water} is the areal extent of water the gas covers and dC is ($C_{sat} - C_w$) where C_{sat} is the saturation concentration in the lake and C_w is the H_2S concentration in the lake. The average plume width over Lake Thingvallavatn during neutral air conditions is about 3 km, based on Gaussian plume theory. The length of the lake in the direction of the plume is about 10 km; thus about 30 km^2 of the lake are covered by the plume. Temperature and pH measurements made in the lake were used for calculating the effective Henry's constant and C_{sat}. The C_w in the lake was assumed to be zero as H_2S is quickly depleted from the water. Table 1 shows the parameters used and calculated. Using the overall transfer velocity reported for pH = 8 by Balls and Liss (1983) for a laboratory experiment, 36.9 cm/h for the equation above and parameters in Table 1 yields an uptake of 70 tons/per year or 0.34% of the H_2S emissions.

The above calculations and analyses of the data indicate that insignificant amounts of H_2S were dissolved in Lake Thingvallavatn compared to the 2012 emissions.

Table 1. Parameters for Lake Thingvallavatn.

Parameter	Value	Reference
Lake pH*	8.5	Eiriksdottir and Gislason (2013)
Lake temperature*	5 °C	Eiriksdottir and Gislason (2013)
k_w	36.9 cm/h	Balls and Liss (1983)
C_{sat}	0.0073 g/m^3	Fernandez-Prini et al. (2003)/ Seinfeld and Pandis (2006)
C_w	0	
A_{water}	30 km^2	

*Mean values found from measured inflow and outflow

4.1.3 Washout

The amount of H_2S that is dissolved in the water that precipitates through the plume was estimated by assuming that the gas and water phase of the H_2S reached equilibrium during the time it takes the precipitation to reach the ground. According to Henry's law the H_2S concentration in the water precipitated, $C_{precip,}$ is

$$C_{precip} = HC_{air} \quad (5)$$

where C_{air} is the H_2S concentration in the plume and H is the dimensionless Henry's constant. Fernandez-Prini et al. (2003) and Seinfeld and Pandis (2006) were used to find the effective Henry's constant at 10°C and at the mean pH of the precipitation in the

area or at pH = 5.6. The method described by Fernandez-Prini et al. (2003) results in the Henry's constant having a unit of pressure, using the ideal gas law to calculate the partial pressure of H_2S in the air yields a molar fraction (x_{H2S}) of dissolved H_2S in the water:

$$x_{H_2S} = \frac{p_{H_2S}}{k^*_{H,H_2S}} = 1.1 \cdot 10^{-10} \quad (6)$$

where p_{H_2S} is the partial pressure of H_2S in the air and k^*_{H,H_2S} is the effective Henry's constant in units of pressure. The total precipitation at Hellisskard Pass in 2012 was 2068 mm, yielding $2.27 \cdot 10^8$ m^3 of water over the plume area. Using the above assumptions, the equation gives about 46 kg of H_2S that can potentially be dissolved in the precipitation. This is only 0.0002% of the total amount of H_2S emitted in 2012, which was negligible. Varying the H_2S concentration and/or the precipitation amounts within a plausible range did not increase the possible washout by any significant amount.

In Figure 5 one hour measurements of H_2S at Station MT are plotted against the hourly precipitation at the station while the wind direction was towards the station from the power plants (84-159°). Gray lines show the 50 and 90 percentiles calculated at 0.5 mm intervals. The figure does not show correlation between the H_2S concentration and precipitation. The highest concentrations however occur while there is no precipitation. This is likely due to the fact that a high H_2S concentration is expected in stable air with little or no cloud cover (Olafsdottir and Gardarsson, 2013, Thorsteinsson et al., 2013). These results may appear to contradict the findings of other studies (Kristmannsdottir et al., 2000, Thorsteinsson et al., 2013) where H_2S concentration was reported to decrease with precipitation. However, the results do not exclude decrease during precipitation but indicate that it was not due to dissolution.

The above calculations and analyses of the data indicate that insignificant amounts of H_2S were washed out with precipitation compared to the 2012 emissions.

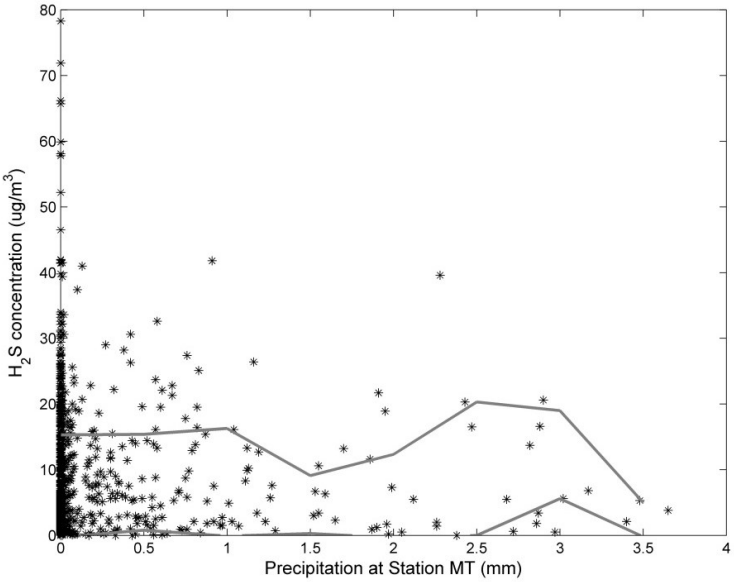

Figure 5. The results of H_2S measurements (one hour averages) plotted against precipitation at Station MT when winds were from the direction of the power plants (84-159°). The solid lines are 50 and 90 percentiles, calculated at 0.5 mm intervals.

4.1.4 Summary of sinks

Table 2 shows a summary of the sinks and associated estimates of H_2S depletion. The table shows that the total of H_2S sinks identified in the study account for about 1.2% of the total H_2S emissions in 2012.

Figure 6 depicts the results in Table 3 showing that oxidation is the largest sink estimated, depleting 143 tons/year, on the basis of the 2012 emission, but leaving the sulfur still in the gas-phase which is likely to be transported rapidly out of the area. Oxidation is indicated to be a slow process, even slower during winter when there is little radiation. The H_2S depleted by other sinks, about 113 tons, are retained in the area, mostly in the surface water of Lake Thingvallavatn or 70 tons/year. Eiriksdottir and Gislason (2013) report an increase of over 1000 tons/year of sulfur in the lake outflow compared to the inflow (when the measured mean sulfur amount in the inflow was extrapolated to the whole inflow), that would equal about 1100 tons/year of H_2S if this difference was because of uptake in the lake. Thus atmospheric uptake does not solely seem to explain the difference between the in- and outflow. About 32 tons/year of H_2S were estimated to accumulate in moss within a few kilometers of the power plants as the accumulation rate decreased exponentially from the source. Moss damage has been confirmed to a greater extent at the NV Power Plant, which has been in operation longer than the HH Power Plant. Whether the damage is due to sulfur accumulation or other processes such as hot water or steam, has not been established. Atmospheric transfer to soils was estimated with Fick's law to be about 9 tons/year which are likely to leach out of the soil (Smith et al., 1972). The estimated uptake potential of the soils was found to be much larger than the estimated uptake as it was limited by the diffusion of the gas through the soil. Washout with precipitation (pH = 5.6) and steam at the source (pH = 6.7) were estimated to be about 46 kg/year and 1.9 tons/year, respectively.

Table 2. Summary of H_2S depletion from the atmosphere by identified sinks.

Atmospheric H_2S depletion	(tons/year)	% of 2012 emission
Oxidation	143	0.69
Washout	0.046	< 0.001
Dissolved in steam	1.9	0.009
Vegetation	32	0.15
Soil	9	0.04
Lake Thingvallavatn	70	0.34
Total	256	1.2%

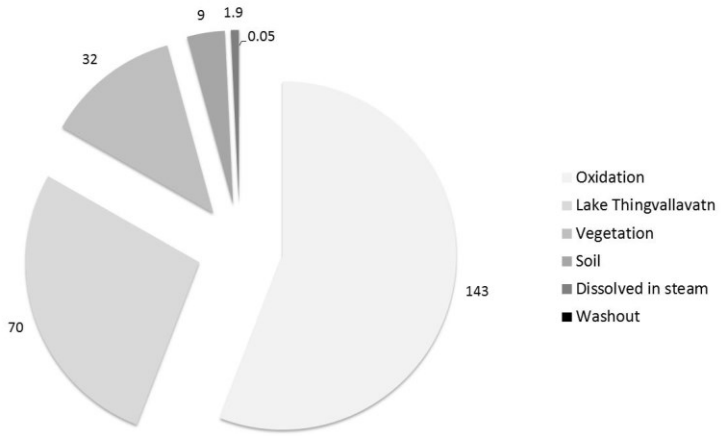

Figure 6. Depletion by each identified sink (tons/year).

4.2 H_2S distribution

4.2.1. Steady state distribution in the study area

Figure 7 shows the study area divided in two main segments, towards the land and towards the ocean. Table 3 shows the yearly amount of H_2S distributed towards each segment based on the 2012 emission data and the percentage of wind observed at Station H (data from 2001-2012) towards each segment. Assuming steady wind conditions and one source at the center of mass of the emissions, about 40% of the hydrogen sulfide was expected to go further inland and about 60% towards the ocean. Some of the H_2S transported over the ocean is likely to be dissolved in the ocean. Most of the H_2S is though likely to be oxidized in the atmosphere over time to SO_2 as indicated by the estimated sinks.

The two smaller segments in Figure 7 include the capital area and Lake Thingvallavatn. The estimation of H_2S sinks showed that the uptake in Lake Thingvallavatn was the second largest sink. The lake is the largest natural lake in Iceland (83 km^2), it is rather deep, cold and surrounded with lava with little vegetation. In spite of this there is a diverse ecosystem in the lake (Jonasson and Hersteinsson, 2002). The lake and its river basin are considered one of Iceland's natural treasures. Table 3 shows that the wind blew towards the lake 10% of the time, yielding about 2070 tons of H_2S crossing the lake and its surroundings per year, based on the 2012 emissions. The estimated uptake is 70 tons/year or about 3.4% of the atmospheric H_2S transported over the lake. As formerly mentioned, there seem to be larger sources of sulfur to the lake other than uptake of atmospheric H_2S.

Based on wind data from Station H, the wind direction was towards the capital area 15% of the time, indicating that about 3100 tons of H_2S were transported across the capital area in 2012. Olafsdottir and Gardarsson (2013) showed that wind direction in which the H_2S concentration at Grensasvegur Measuring Station increased lasted over 30% of the time. This difference indicates effects of terrain and shows in part the difference in estimating one main source instead of two. This indicates that the amount of H_2S travelling towards the capital area might be somewhat underestimated.

Table 3. Segment division of the study area.

Segment	Angle size degree	Size (km^2)	Wind direction towards segment (°)	Wind towards segment (%)	H_2S (1,000 tons/year)*
Lake Thingvallavatn	50	535	200-250	10	2,068
Capital area	50	535	80-130	15	3,103
Land	180	1924	130-310	41	8,480
Ocean	180	1924	310-130	59	12,204

*Based on 2012 emission data

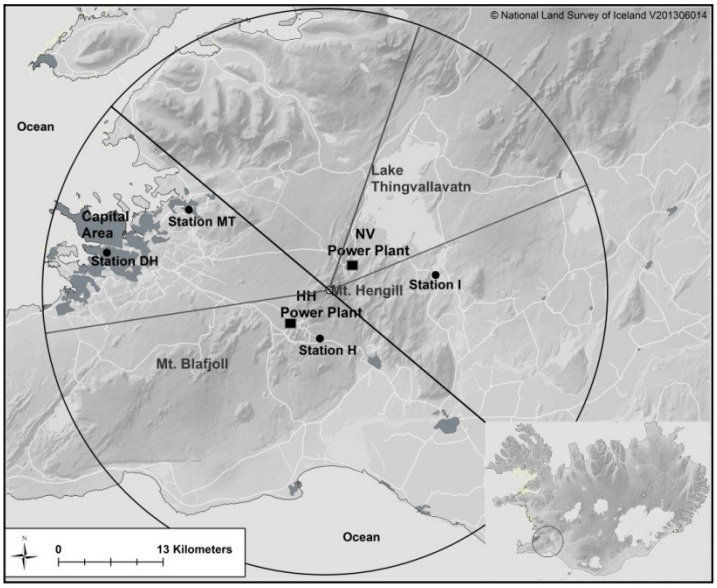

Figure 7. Segment division of the study area.

4.2.2. Distribution in complex terrain

It is indicated that assuming steady wind conditions in the study area is an oversimplification. Measurements along major highways in the area showed that distribution was dependent on spatial variations in wind field and terrain (Olafsdottir et al., 2014b). Figure 8 shows the measurements during five events on Road 39 (Figure 4). All events were measured during neutral or stable air conditions. During events I and II the concentrations were low at first but rose after about 6-8 km on the road consistent with a mountain pass south of the HH Power Plant, indicating that the plume path is through the pass (Path I in Figure 9). During events III-V concentrations decreased within the first 5 km on the road but were measured further on Road 417 (Figure 4 and Figure 9), when compared to meteorological data the plume was shown to deviate from the original path as shown in Figure 9, Path II (Olafsdottir et al., 2014b). Furthermore, the concentration was shown to be relatively high and steady along the mountain in some events indicating trapping of the plume. The distribution of the H_2S has thus been shown to be dependent on terrain features and variations in the wind field.

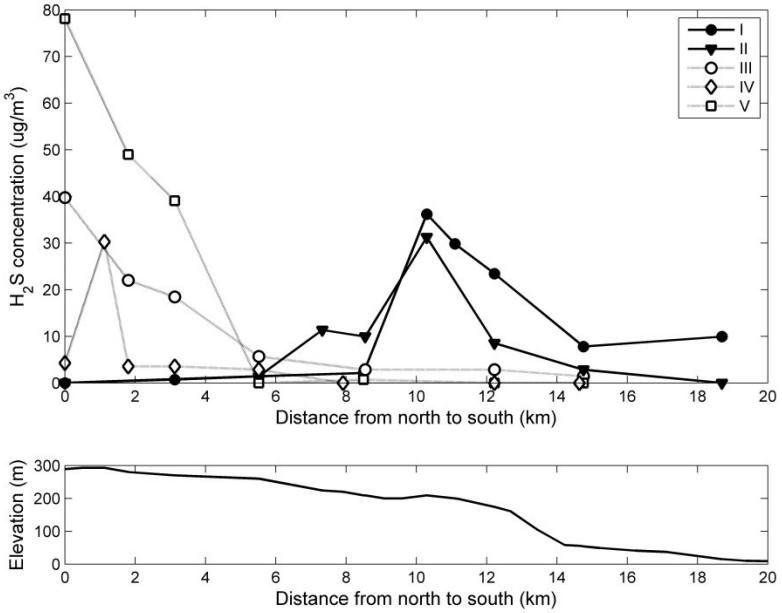

Figure 8. Upper panel: Measured H_2S along Road 39 (Xs1 and Xs2 in Figure 4 are 0 and 23 km on the x-axis, respectively). Lower panel: Elevation of Road 39 from Xs1 to Xs2 in Figure 4.

4. CONCLUSIONS

The results show that natural near field sinks of atmospheric H_2S within 35 km from the NV and HH Power Plants are insignificant compared to the emissions, with about 99% of the H_2S being transported out of the area as H_2S. About 143 tons/year are estimated

to be oxidized within the area but being quickly removed with wind while 113 tons/year are estimated to be retained in the area in surface water, vegetation and soil. There are large uncertainties in the estimation but the estimates are arrived at in a conservative way, that is, the number should represent an approximate upper bound. Plumes of H_2S were shown to follow depressions in the terrain and accumulate by a mountain range during neutral and stable air.

The results of the research show that the distribution of H_2S from the NV and HH Power Plants was affected by wind and terrain, often in narrow plumes, and that it may accumulate along mountain ranges or in lows in the landscape but in the end, most of it is transported out of the area as H_2S. The sinks estimated indicate that the H_2S is oxidized to SO_2 with time in the atmosphere or dissolved in the ocean. Other sinks are expected to be insignificant.

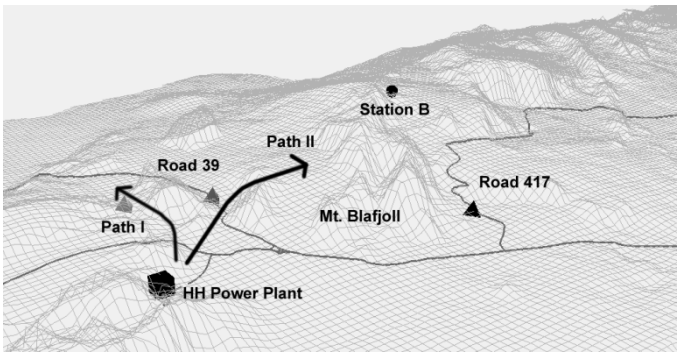

Figure 9. View from northeast to southwest towards the sea, looking south from the HH Power Plant. Estimated plume paths are shown.

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