EMISSION, DISPERSION AND REACTION OF H₂S IN STEAM FROM GEOTHERMAL FIELDS IN ICELAND

Hrefna KRISTMANNSDÓTTIR*, Magnús SIGURGEIRSSON*, Halldór ÁRMANNSSON* and Hreinn HJARTARSON+

*Orkustofnun, Grensásvegur 9, 108 Reykjavík, +The Icelandic Meteorological Institute, Bústaðavegi 9, 105 Reykjavík.

ABSTRACT

Gas emissions from geothermal fields in Iceland have been studied within the scope of a project aimed to enhance research concerning environmental aspects of geothermal exploitation. Short time measurements of the gases have been carried out in several high-temperature geothermal fields in Iceland. In four exploited fields baseline values of the concentration of sulphur gases, H₂S and SO₂ have been obtained by long term measurements. The data reflect the depency between gas concentration and weather conditions very strongly. Preliminary interpretation and modeling indicate minor or very slow conversion of H₂S to SO₂ at atmospheric conditions.

INTRODUCTION

As a consequence of the increased emphasis on the environmental viability of energy projects a project was started in Iceland to study the environmental impact of geothermal exploitation. The project was initated by Orkustofnun, which requested the cooperation of the main exploiters of high-temperature geothermal energy in Iceland. The project entailed firstly an assessment of the present status at the five main sites at high-temperature geothermal exploitation in Iceland and secondly the definition of several priority projects (Kristmannsdóttir and Ármannsson, 1995) to be carried out within the scope of the project.

One of the main effects on the environment of geothermal exploitation is the emission of gases in the geothermal steam (Axtman, 1975, Ármannsson and Kristmannsdóttir, 1992). The greenhouse gases CO₂ and CH₄ are of main concern in this respect, along with sulphur gases. The sulphur gas emitted from geothermal plants is in the form of H₂S. As this gas is toxic in high concentrations and has a very unpleasant smell in low concentrations its presence is often of great environmental concern. It is however, only inside poorly ventilated buildings at plant or drill sites, that people are in any danger of being poisoned. As unpleasant as the smell may be to people not used to it people accustomed to it may not notice it at all. The H₂S gas may however be oxidized to SO₂ that causes acidification of rain which is of great concern and globally watched. If all the H₂S gas was to be quantitatively converted to SO₂, at least some geothermal power plants would be considered rather polluting and the removal of H₂S from steam made mandatory. Measurements of pH and sulphate concentrations in precipitation in the vicinity of

the Olkaria power plant in Kenya (Muna and Ojambo, 1985) and of pH in the neighbourhood of the Svartsengi plant (Bjarnason, 1991), reaveal neither pH changes nor SO₂ addition. There is evidence that not all the H₂S is converted to SO₂ and anyway not immediately. This is a matter of considerable discussion as the sulphur chemistry is quite complicated and this question has not been the topic of much researh sofar. Some of the H₂S is apparently oxidized to sulphur and accumulates near or within the geothermal field. The solid sulphur precipitated will gradually react with the soil to form metal sulphates and may be beneficial rather than harmful to the environment. In the litterature there are conflicting views about oxidation of H₂S in the atmosphere. Brown and Webster (1994) claim that oxidation of H₂S within aerosols is rather a slow process. Cox and Sandalls (1974) claim that photo-oxidation of H₂S to SO₂ is a major loss process for H₂S in atmosphereic air.

Sulphur gas emissions are thus first a local pollution concern as requirements posed by environmental authorities need to be met. Secondly there is global pollution concern because of international conventions regarding emission of SO₂ into the atmosphere. In that respect the question about conversion of H₂S to SO₂ is of major concern.

In 1991 when the environmental research program was initiated at the National Energy Authority in cooperation with the main companies exploiting geothermal steam in Iceland, little data was available on the concentration and dispersion of H₂S in the atmosphere within and outside the geothermal fields. Thus one of the main tasks within the project was to measure the concentration of sulphur gases in the atmospere within and outside the sites of geothermal power plants.

To study the conversion of H_2S to SO_2 similar monitoring stations were also established at some distance outside one of the fields to obtain concentration values for the atmosphere. Those values are used as a basis for modeling the dispersion of the gases in the atmospheric air. Some simple experiments are also underway to study the physical conditions and time needed to convert H_2S to SO_2 .

EMISSION OF H₂S FROM ICELANDIC GEOTHERMAL FIELDS

A total of about 14.000 tons of H₂S is estimated to be emitted yearly from geothermal areas in Iceland (Fig. 1). Of those about 8.000 tons are emitted from geothermal power plants. In Table 1 and Fig. 2 the estimated outflow from all the exploited areas are shown. In Fig. 2 the estimated outflow from selected non producing fields is shown for comparison.

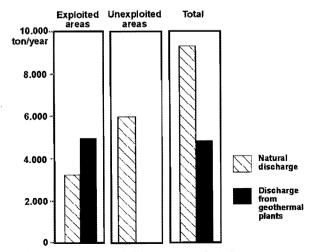


Figure 1. Annual discharge of H₂S from Icelandic geothermal fields.

Table 1. H₂S emission from exploited geothermal fields in Iceland

Geothermal field	H ₂ S emission	in tons/year	
	from fumaroles	from drillholes	Total
REYKJANES	5	60	65
SVARTSENGI	15	170	185
NESJAVELLIR	140	5200	5340
NÁMAFJALL	470	1300	1770
KRAFLA	700	1600	2300
HVERAGERÐI	140	40	180

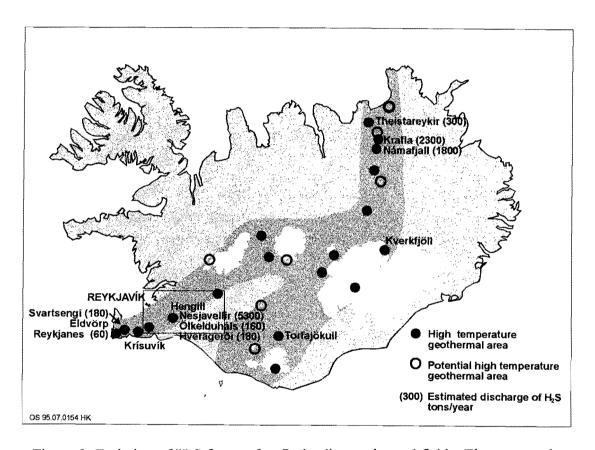


Figure 2. Emission of H_2S from a few Icelandic geothermal fields. The rectangular area shows the location of the map in Fig.5.

The Reykjanes and Svartsengi fields are brine fields with relatively low H_2S concentrations. The steam production from Reykjanes is also as yet rather small. In Námafjall there is a low production, but high concentration of H_2S in the steam. In the Krafla field production is much higher than in Námafjall, but the H_2S concentration of the steam is considerably lower. In the Nesjavellir field there is both high production and high H_2S concentration of the steam and the main part of the H_2S emission in Iceland comes from that area.

PRELIMINARY MEASUREMENTS OF THE CONCENTRATION OF SULPHUR GASES IN THE ATMOSPHERE

In 1993 preliminary measurements were made of the concentration of sulphur gasses and mercury in the atmosphere within ten high-temperature geothermal fields in Iceland, producing and non-producing (Ivarsson et al. 1993). The aim of the study was partly to get a rough estimate of the concentration levels of the gases and partly to compare the precision, accuracy and usefulness of several different analytical methods. The measurements were both point measurements and measurements based on sampling of gases over 24 hours in wetted filters and liquids. The range in H₂S concentration was found to be from <1 to >200 µg/m³ and the variation within the same area could be almost of the same order of magnitude from one time to another. The highest concentration was as expected found in the Nesjavellir field, about 40 times that from the Svartsengi field. The range in SO₂ concentration was from <0.1 to about 18 µg/m³ also with great variations from one time to another at the same place. As the weather in Iceland and therefore the wind directions are quite changable the great periodic variations observed were not unexpected. The concentration of mercury in the atmosphere was found to be of the same order as over the oceans in the northern hemisphere and confirmed the results obtained during a previous study by LIDAR technique (Edner et. al, 1991).

LONG TERM MEASUREMENTS OF THE CONCENTRATION OF SULPHUR GASES WITHIN GEOTHERMAL FIELDS

The results of the short time measurements of the concentration of sulphur gasses in the atmosphere showed that to get reliable baselines for the concentration levels measurements had to be carried out over several months.

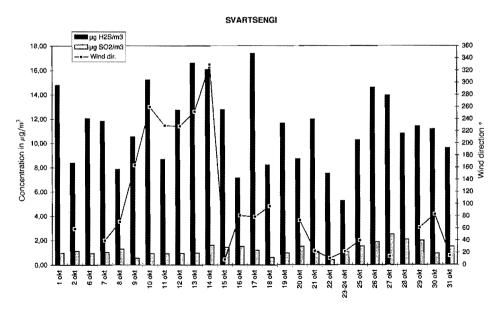


Figure 3. Measurements of H₂S and SO₂ over one month in the Svartsengi field. Wind directions are shown to the right.

Method testing showed that the method based on sampling on filters wetted in KOH and AgNO₃ solutions was most reliable and therefore chosen for further work. The filters were changed daily and the sampling was continued through 4-6 months in four of the producing high-temperature geothermal fields: Svartsengi, Nesiavellir, Krafla and Námafjall.

The long term measurements were carried out over the years 1994-1996 Sigurgeirsson and Kristmannsdóttir, 1995, 1996a, 1996b). The dependency of the H₂S concentration on weather is still significant in the long term measurements, both on wind direction (Fig. 3) and precipitation (Fig. 4). As the wind is very changable in Iceland one would not expect the dependency on wind direction to be reflected in the mean concentration for each day. This correlation is clear on some days, but not at all on other days. When there is heavy rain or snow H₂S appears to be almost quantitatively eliminated, probably due to dissolution in the precipitation. No clear dependency of the SO₂ concentration on weather conditions is observed. No direct correlation between the concentration of the two sulphur gases has been found.

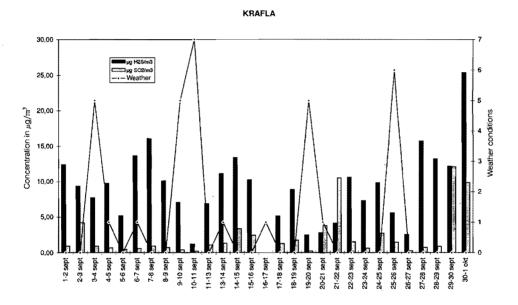


Figure 4. Measurements of H₂S and SO₂ over one month in the Krafla field. Weather conditions are indicated by the numbers to the right: 1-3 are days with no precipitation, 4 means foggy days, 5 a little rain, 6 heavy rain.

The mean concentration (Table 2) obtained for the four places are not so different for H₂S. They are slightly higher for Nesjavellir than for the others and Svartsengi is slightly lower than Krafla and Námafjall. The mean concentration for SO₂ varies more. It is lowest in Svartsengi and there the concentrations are not very different in summer and winter. At Nesjavellir the mean concentration is higher than in Svartsengi, similar though during the summer, but higher during the winter. In Krafla the values are generally higher than both in Svartsengi and Nesjavellir. In Námafjall the values are considerably higher than in the other fields. At a diatomite factory operating within the geothermal field there is burnt crude oil, which is responsible for the air contamination. This may also affect the the values from Krafla, which is 11 km from Námafjall.

Table 2. The mean concentration of H_2S and SO_2 in the atmosphere within and outside geothermal fields.

Location	Mean conc. of H ₂ S in μg/m ³	•	Mean conc. of SO ₂ in μg/m ³	The range of conc. of SO ₂ in µg/m ³
Nesjavellir	13	9.5-15.2	1.7	1.0-2.9
Svartsengi	10	6.6-11.6	1.0	0.8-1.3
Krafla	11	9.6-12.1	2.4	2.1-2.8
Námafjall	11	10.5-11.4	5.5	2.5-8.3
Írafoss	1.0	0.4-2.0	0.1	0.1-0.2
Korpa	0.5	0.3-1.0	0.4	0.3-0.5
Reykjavík	_	_	1.6	0.4-2

⁻ Not measured

MEASUREMENTS OF THE CONCENTRATION OF SULPHUR GASES IN TWO MONITORING STATIONS OUTSIDE NESJAVELLIR

Long term measurements were carried out at two sampling points in at a distance from the Nesjavellir field (Sigurgeirsson and Kristmannsdóttir, 1996b) in order to study the dispersion and possible conversion of H₂S to SO₂ by time. The sampling points, Korpa and Írafoss are shown in figure 5.

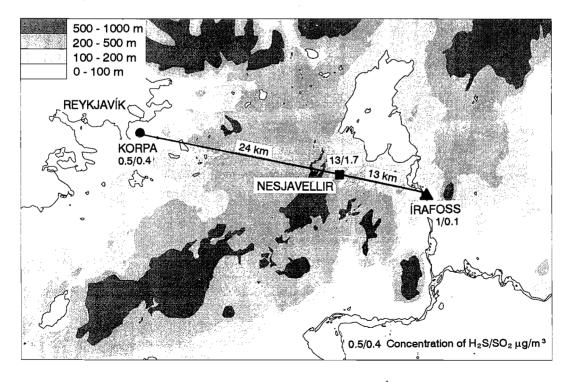


Figure 5. The location of the gas monitoring stations at Írafoss and Korpa, relative to the Nesjavellir geothermal field. The location of this map within Iceland is shown in Figure 2.

The measurements were run continously for one year at those points. The mean concentrations are shown in table 2. In the table the annual mean concentration of SO_2 (Benjamínsson, 1993) is also shown for comparation. The concentration of H_2S is higher at Írafoss than at Korpa, but the reverse is true for SO_2 .

Korpa is nearer to the town of Reykjavík than both Nesjavellir and Írafoss and pollution from transportation and industry would be expected to influence the concentration of SO₂. From Nesjavellir to Korpa there is shorter distance than from Nesjavellir to Írafoss, but there are high mountains blocking the way. In spite of longer distance, the way from Nesjavellir to Írafoss would be expected to be much less influenced by the landscape. Most of the H₂S both at Korpa and Írafoss is expected to have originated from Nesjavellir. No measurements have been made of H₂S in the atmosphere in the town of Reykjavík.

DISCUSSION AND CONCLUSION

Concern about sulphur gas emissions during geothermal production is partly due to potential local pollution and partly to global environmental requirements regarding emission of SO₂ into the atmosphere. Measurements of the concentration of sulphur gases in air within the geothermal fields show great temporal variation. As expected the concentration is very much related to weather, both wind direction and precipition.

The mean concentration of H₂S measured over several months is surprisingly similar in all the production fields. In Nesjavellir where it is highest it is still of the same magnitude as in Svartsengi whereas the amount of H₂S emission is 30 times less.

The concentration of SO_2 in the atmospheric air within the geothermal fields varies more than that of H_2S . The reason for this difference has been shown to be mainly due to variable pollution from factories and transport. High concentrations at Námafjall and probably also in Krafla are caused by pollution from the burning of crude oil at a diatomite factory. Good background values for H_2S and SO_2 in the atmospheric air in Iceland outside towns are not available, but the concentration of SO_2 is normally below $0.1~\mu g/m^3$ and that of H_2S is lower than $0.3~\mu g/m^3$ (detection limit for the human nose).

The concentration of SO₂ in the atmospheric air at Nesjavellir is almost the same as within the town of Reykjavík, whereas at Korpa the concentration is about one fourth of the Reykjavík value. As Korpa is much nearer to Reykjavík than Nesjavellir the pollution from the town should be much more prominent there. As no other polluting factors are found in the vicinity of Nesjavellir the excess SO₂ of Nesjavellir is probably due to conversion of H₂S to SO₂. The SO₂ concentration is still very low compared to that of H₂S and the total amount of H₂S emitted from wells and fumaroles. Similar argument applies to Svartsengi.

How much of the H_2S will eventually be converted to SO_2 is still somewhat uncertain. Only a small fraction of the H_2S appears to be converted to SO_2 within the fields. This is found to be true even on days with almost no wind when the emitted gas will stay within the fields for a while. On days of abundant precipitation H_2S appears to be almost quantitatively washed out.

The monitoring stations at Korpa and Írafoss were established to obtain values for use as a basis to model the dispersion of the gasses. The modelling work is underway, but suffers because the data is not very plentiful. There are some ambiguities in the measurements results. The SO₂ concentration at Írafoss is almost at background level, whereas the higher one at Korpa is surely to some extent due to pollution from Reykjavík.

At present it appears that H₂S is converted so slowly to SO₂ that only a fraction of it reacts within the grothermal fields. As humidity will effectively wash out H₂S from air only a fraction of the H₂S dispersed away from the geothermal is believed to end up as SO₂. Work is underway to quantfy this fraction. Simple experiments to study the physical conditions and time needed to convert H₂S to SO₂ are in progress. Together with results of modeling work they will probably yield some quantitative estimate of the amount of H₂S converted to SO₂ on average.

REFERENCES

Axtmann, R.C., 1975. Emission control of gas effluents from geothermal power plants. *Environmental Letters*, 8, 135-146.

Ármannsson, H. and Kristmannsdóttir, H. 1992. Geothermal environmental impact. *Geothermics*, 21, 869-880.

Benjamínsson, J., 1993. "Pollution of air in the town of Reykjavík 1993". A yearly report from the Health Authorities in Reykjavík (in Icelandic).

Bjarnason, J. Ö., 1991. On the pH of precipitation in Svartsengi. Orkustofnun Report (in Icelandic), JÖB-91/02.

Brown, K. L. and Webster, J.G., 1994. H₂S oxidation in aerosols. *Proceedings of the 15th PNOC-EDC Geothermal Conference*, Mahati Philippines, 37-44.

Cox, R.A. and Sandells F.J., 1974. The photo-oxidation of hydrogen sulphide and dimethyl sulphide in air. *Atmospheric Environment*, 8, 1269-1281.

Edner, H., Faris, G. W., Suneson, A., and Swanberg, S., Bjarnason, J. Ö., Kristmannsdóttir, H. and Sigurðsson, K.H., 1991. Lidar search for atmospheric atomic mercury in Icelandic geothermal fields. *J. Geophys. Res.*, 96, 2977-2986.

Ívarsson, G., Sigurgeirsson, M.Á., Gunnlaugsson, E., Sigurðsson, K.H. and Kristmannsdóttir, H 1993. Measurements of gas in atmospheric air. The concentration of hydrogen sulphide, sulphur dioxide and mercury in high-temperature geothermal areas. A co-operative project of Orkustofnun and Hitaveita Reykjavíkur (in Icelandic). Orkustofnun OS-93074/JHD-16, 69 p.

Jónsson, V.K., Abrahamson, D. and Sólnes, J. (1991). The emission of greenhouse gases in Iceland and means of improvement (In Icelandic). In *Orkuthing 91*, Ó.B. Smárason (Ed.), Stapi, Reykjavík, 237-253.

Kristmannsdóttir, H. and Ármannsson, H. 1995. Environmental impact of geothermal utilization in Iceland. *Proceedings of the World Geothermal Congress* 1995, Florence, Italy, 2731-2734.

Muna, E. W. and Ojambo, S. B., 1985. Possible influence of gases emitted from Olkaria geothermal field on the rainwater of its surroundings. Kenya Power Company Ltd., Geothermal Project, Olkaria. Report No. GC/GEN/038.

Sigurgeirsson M. Á. and Kristmannsdóttir, H., 1995. "Measurements of sulphur gasses in the atmosphere. The concentration of hydrogen sulphide and sulphur dioxide at Svartsengi and Krafla. Orkustofnun Report (in Icelandic) OS-95025/JHD-18B.

Sigurgeirsson M. Á. and Kristmannsdóttir, H., 1996 (a)."Measurements of sulphur gasses in the atmosphere. The concentration of hydrogen sulphide and sulphur dioxide at Nesjavellir and in Bjarnarflag". Orkustofnun Report (in Icelandic) OS-96020/JHD-10B.

Sigurgeirsson M. Á. and Kristmannsdóttir, H., 1996 (b) "Measurements of sulphur gasses in the atmosphere. The concentration of hydrogen sulphide and sulphur dioxide at Korpa and Írafoss". Orkustofnun Report (in Icelandic) OS-96020/JHD-11B.