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Edner, H; Faris, G. W; Sunesson, A; Svanberg, Sune

Published in:

Journal of Geophysical Research

DOI:

[10.1029/90JD02350](https://doi.org/10.1029/90JD02350)

1991

[Link to publication](#)

Citation for published version (APA):

Edner, H., Faris, G. W., Sunesson, A., & Svanberg, S. (1991). LIDAR Search for Atmospheric Atomic Mercury in Icelandic Geothermal Fields. *Journal of Geophysical Research*, 96, 2977-2986.
<https://doi.org/10.1029/90JD02350>

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4

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LUND UNIVERSITY

PO Box 117
221 00 Lund
+46 46-222 00 00

Lidar Search for Atmospheric Atomic Mercury in Icelandic Geothermal Fields

H. EDNER, G. W. FARIS, A. SUNESSON, AND S. SVANBERG

Department of Physics, Lund Institute of Technology, Lund, Sweden

J. Ö. BJARNASON, H. KRISTMANNSDÓTTIR, AND K. H. SIGURDSSON

Geothermal Division, Orkustofnun, Reykjavik

A search for atmospheric atomic mercury as a possible tracer gas for geothermal energy exploration was performed in three Icelandic geothermal fields using the differential absorption lidar technique. Contrary to expectations, concentrations basically only at the Atlantic background value of about 2 ng/m³ were found in Iceland.

INTRODUCTION

Atomic mercury is known to be a tracer gas in connection with a variety of geophysical phenomena, such as ore deposits [Bristow and Jonasson, 1972; McCarthy, 1972; Kromer *et al.*, 1980] and geothermal [Varekamp and Buseck, 1983; Robertson *et al.*, 1977], seismic [Fursov *et al.*, 1968] and volcanic [Varekamp and Buseck, 1981; Olafsson, 1975] activities. The present paper describes laser remote sensing measurements performed in Icelandic geothermal fields in order to assess the possibility of using atmospheric atomic mercury anomalies for geothermal prospecting. Previous independent studies with point monitors at the Geysers geothermal plant in California and at the Cerro Prieto plant in Mexico revealed high concentrations of atomic mercury in noncondensable gases in the steam emerging from the well-heads (300–5,800 µg/m³). Mercury was also measured in the cooling tower exhaust air at a much lower level (0.18–0.83 µg/m³) due to extremely large gas dilution with ambient air [Robertson *et al.*, 1977]. Mercury was measured by first sampling gas through selective absorbents followed by volatilization and flameless atomic absorption analysis (AAS). Sixty to ninety percent of the collected mercury was considered to be in the form of atoms, Hg⁰.

In independent measurements at the Geysers geothermal plant a multipass atomic absorption spectrometer sensitive only to atomic mercury was used [Jepsen, 1973]. In one geothermal plume more than 28 µg/m³ was detected while the concentration in ambient air at the field site was of the order of hundreds of nanograms per cubic meter.

Geothermal energy is important in Iceland, where about 85% of the energy for domestic heating is extracted from geothermal installations. An extensive program for exploration and characterization of geothermal fields is being pursued. Chemical analysis of the geothermal water, steam, and gas is performed for most active wells in high-temperature fields. [see Kristmannsdóttir, 1984]. Mercury concentrations have been determined, but the techniques used do not differentiate between different oxidation states of mercury. The samples are collected into a permanganate solution

acidified with nitric acid and analyzed by flameless AAS after amalgamation in gold [Olafsson, 1974]. Total mercury concentrations in well fluids in Icelandic high-temperature geothermal fields range up to 7000 ng/L. However, what fraction is atomic mercury was unknown. The present paper addresses this question.

Recently, the differential absorption lidar (DIAL) technique [Killinger and Mooradian, 1983; Measures, 1984] has been extended to measurements of atmospheric Hg (atoms) [Aldén *et al.*, 1982; Edner *et al.*, 1989]. The natural background concentration of Hg is about 1–2 ng/m³ over oceans [Slemr *et al.*, 1981] and somewhat higher over continents. A dominant fraction of this mercury is in atomic form. By using integration over an optical pathlength of about 1 km the DIAL technique operating at the mercury absorption line close to 254 nm is capable of measuring concentrations down to this exceedingly low level. Range-resolved measurements over smaller subsequent integration intervals can be performed for correspondingly higher concentrations.

The Hg DIAL technique, being a remote sensing technique, is of particular interest for geophysical surveys, since it has the potential of covering large areas, particularly if implemented in an airborne version. Aerosol backscattering can be used for range-resolved measurements while echoes of topographic targets, such as the ground, can be utilized for path-averaged measurements. For airborne applications using topographic targets, the gas correlation version of the lidar technique [Edner *et al.*, 1984] may be especially well suited.

A particular feature of the Hg DIAL method, being an *in situ* spectroscopic technique, is that it is sensitive only to atomic mercury and not to chemical compounds of mercury. The DIAL remote sensing technique shares this feature with multipass absorption spectrometers with background rejection by gas filter techniques or by Zeeman modulation [Robbins, 1973]. Such instruments are point monitoring devices also operating on unprepared ambient air. In contrast, sampling and trapping techniques require great care to allow an unambiguous determination of the oxidation state of mercury. Measurements with a Hg absorption spectrometer as discussed above [Jepsen, 1973; Robbins, 1973] definitely reveal elevated air concentrations of atomic Hg in connection with certain mines.

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Paper number 90JD02350.
0148-0227/91/90JD-02350\$05.00

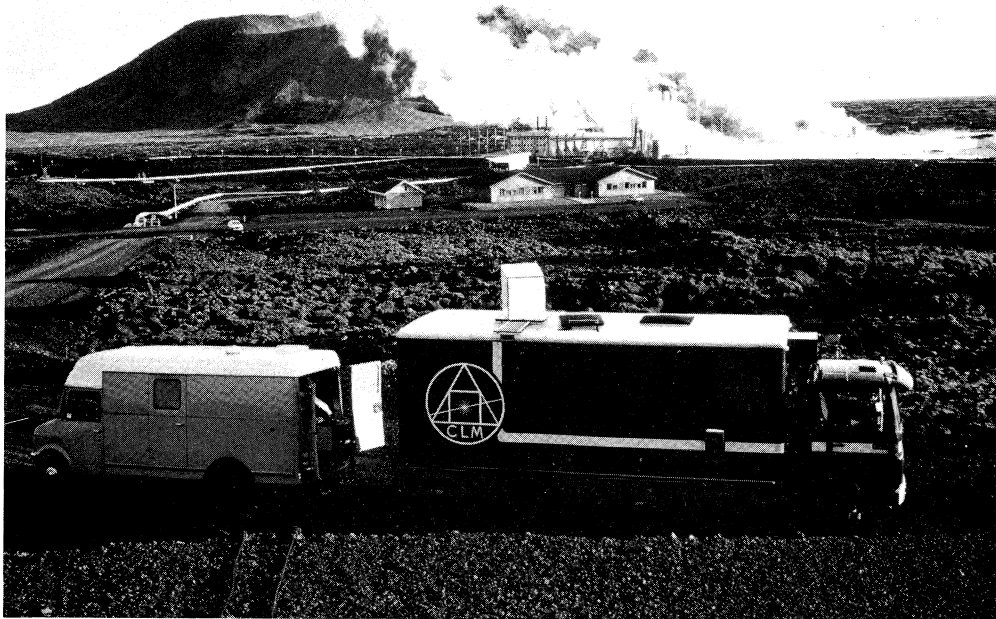


Fig. 1. Lidar system and supporting vehicle photographed at the Svartsengi geothermal heat and power plant, Iceland.

With the background knowledge presented it was judged to be interesting to investigate with the lidar technique if elevated concentrations of atomic mercury could be found in association with Icelandic geothermal fields.

In the next section the Hg lidar system used in the reported field experiments is briefly described, as is a multipass optical absorption setup used both in the field and in laboratory measurements on extracted well gas samples. Then the measurements performed at different field locations are described. Finally, the outcome of the field experiments, yielding exceedingly low atomic Hg concentrations, is discussed.

Hg LIDAR SYSTEM DESCRIPTION

Measurements were performed in three different Icelandic geothermal fields, each of different character. A newly constructed Swedish mobile lidar laboratory and a supporting laboratory van containing a large multipass optical absorption cell (White cell) and other equipment were shipped to Iceland by a container vessel. The vehicles are shown in the Icelandic lava field environment in Figure 1. The construction of the lidar system and measurement examples have been given by *Edner et al.* [1987]. The system has been further refined to allow atomic mercury monitoring, and the first successful mapping of Hg plumes has been reported in measurements by this group at a Swedish chlorine-alkali plant [*Edner et al.*, 1989]. Since these two papers give a full account of the techniques used we will here only give a brief description.

The laser source is a Nd:YAG system pumping a tunable dye laser. The dye laser is operated at wavelengths around 507 nm with Coumarine 500 dye. The pulse energies are 25–30 mJ, and the pulse repetition rate is 10 Hz. The dye laser output is frequency doubled to the UV region by using a beta barium borate (BBO) crystal (CSK Company, Los

Angeles, California), yielding pulse energies up to 5 mJ at 254 nm. The dye laser wavelength is calibrated by separating 10% of the frequency-doubled beam and by monitoring the absorption of this beam in a cell of mercury vapor. In the measurements repeated control of the proper wavelength setting of the dye laser is performed using the mercury cell. In the DIAL measurements the laser is tuned on and off resonance with the mercury line every second laser shot.

The UV beam is directed into the atmosphere by two right angle quartz prisms and a large plane mirror. The mirror can be rotated around the horizontal axis and the vertical axis, thus determining the direction of the output beam. Backscattered radiation is received by the same mirror and directed to a 40-cm-diameter Newtonian telescope. An interference filter selects the appropriate wavelength range, and a photomultiplier detects the radiation. In order to reduce the requirements of dynamic range in the detection electronics, the photomultiplier gain was ramped up to reach its full value at a range of about 400 m.

A transient recorder performs A/D conversion of the signal with a time resolution of 10 ns, giving a range resolution of about 1 m. The digital signals are averaged on a computer that subsequently stores them on floppy disks. During a measurement the computer controls laser firing, dye laser wavelength setting, beam direction, and data acquisition.

For Hg measurements, the resonance line at 254 nm is used. The laser radiation is tuned on and off the resonance and the "on" and "off" signals are stored separately. By calculating the ratio of the on and off signals the absorption from Hg at a certain distance can be monitored, and the concentration as a function of distance can be calculated. By measuring in several directions through a plume containing Hg, concentration mapping of the plume can be obtained.

As an illustration of the system performance a mapping of

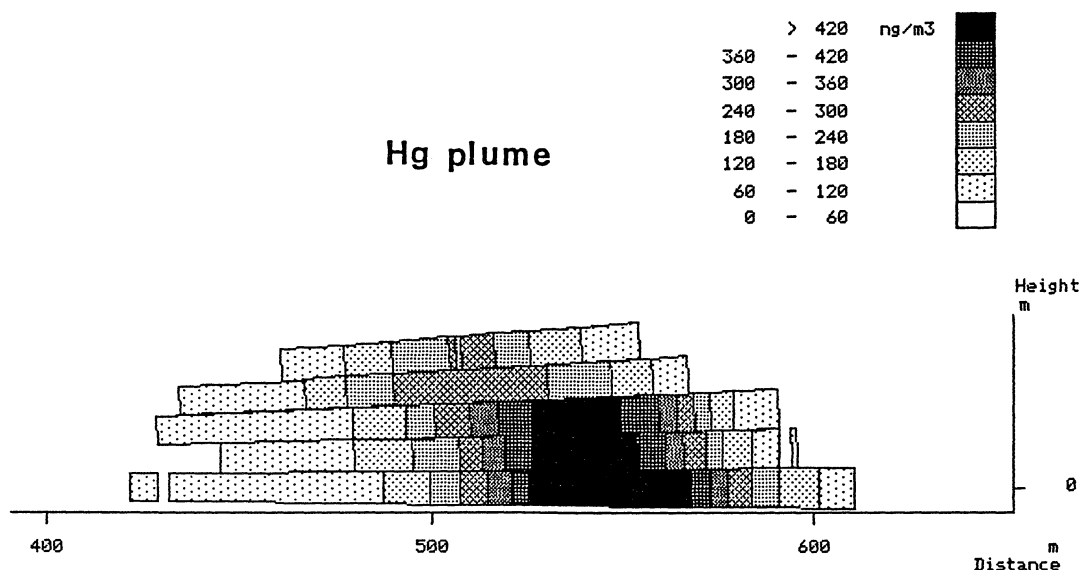


Fig. 2. Computer readout of the Hg atomic concentrations in a vertical slice cut through the plume from a Swedish chlorine-alkali plant.

the Hg plume from a chemical plant is given in Figure 2. Maximum concentrations in the plume are about 500 ng/m^3 . A 50-m integration interval yields a reasonable plume mapping for 10 times lower concentrations as illustrated in the figure. By multiplying the concentration value integrated over the plume cross section by the wind velocity perpendicular to the measured section a total flux of about 20 g/h can be calculated. Background concentrations down to the 2 ng/m^3 level can be measured by increasing the integration length to 1 km as illustrated in our earlier paper [Edner *et al.*, 1989]. Thus, the equipment is well suited to the detection of any Hg anomalies present in the air of the geothermal fields.

MEASUREMENTS AND RESULTS

Field measurements were performed in three geothermal areas: Svartsengi, Krysuvik, and Nesjavellir. These sites are indicated on the map shown in Figure 3.

Svartsengi

The Svartsengi geothermal field is located in a fissure swarm on the Reykjanes peninsula in southwestern Iceland. The peninsula, which consists of lava flows and hyaloclastite formations, is the landward continuation of the Mid-Atlantic Ridge. The Svartsengi geothermal reservoir, classified as liquid-dominated, is highly fractured and very permeable. There are few natural surface manifestations of geothermal activity in the field.

The geothermal fluid is a mixture of approximately 65% seawater and 35% fresh water, at a temperature of about 240°C . Reactions with the surrounding rock have altered the composition of this mixture. Accordingly, the fluid contains approximately 12,700 mg/kg chloride, 6400 mg/kg sodium, 1060 mg/kg potassium, 985 mg/kg calcium, and 430 mg/kg silica. The retrograde solubilities of calcium sulfate and of magnesium silicates have depressed the concentration of sulfate roughly sixtyfold, to about 30 mg/kg, and that of magnesium a thousandfold, to about 0.80 mg/kg. The fluid furthermore contains dissolved gases, primarily carbon di-

oxide and hydrogen sulfide, which are present in amounts of the order of 400 mg/kg and 6 mg/kg, respectively.

The Svartsengi geothermal district heating and power plant (Figure 1) provides a population of 20,000 in the nearby communities with hot water for space heating and domestic use. In addition, the plant produces 8 MW of electric power. Six wells, ranging in depth from 1000 m to 2000 m, supply the plant with a mixture of water and steam. The flow rate of each well is about 60 kg/s. In the plant, the mixture is separated at 156°C (5.5 bar absolute); the steam is used for electric power generation, and the gases, which partition overwhelmingly into the steam phase, are vented to the atmosphere. The brine phase is used to heat cold fresh water, by an arrangement involving heat exchangers as well as a second stage of flashing, this one at 70°C . The fresh water is then piped to consumers, and the spent brine is discharged to a pond.

When samples of the well fluids are collected, the steam-water mixture is separated at the wellhead pressure of 16.3 bar absolute. Under these conditions, the mass fraction of steam is roughly 0.09. Analyses of mercury by the flameless AAS method yield approximately 4300 ng/kg for steam phase, including gases, and about 80 ng/kg for the liquid. This corresponds to a concentration of 460 ng/kg in the single-phase downhole fluid.

A seventh, shallow (425 m) well supplies some 5 kg/s of dry steam, at a temperature of approximately 223°C . Mercury concentrations in this steam were found to range as high as 18,500 ng/kg.

The mercury concentration in ambient air in Svartsengi, at ground level near the power plant, was found, by the same method (flameless AAS), to be about 5 ng/m^3 .

Many different types of test measurements were performed with the lidar system positioned as illustrated in Figure 1. An example of a DIAL measurement through the power plant plume is given in Figure 4. The laser beam was directed to a point where most of the visible water vapor plume had evaporated. In the upper part of the figure on- and

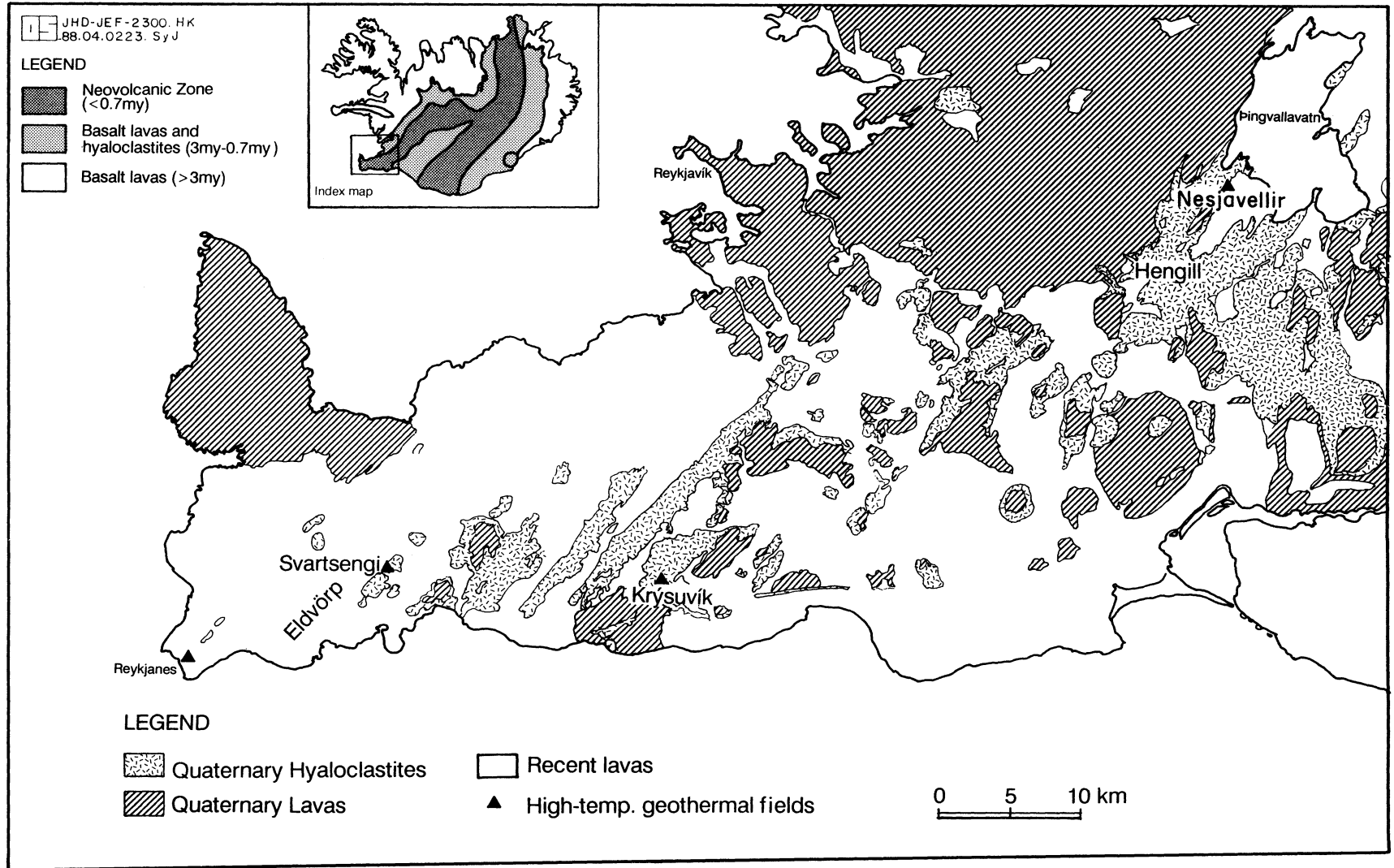


Fig. 3. Map of southwest Iceland with visited geothermal fields indicated.

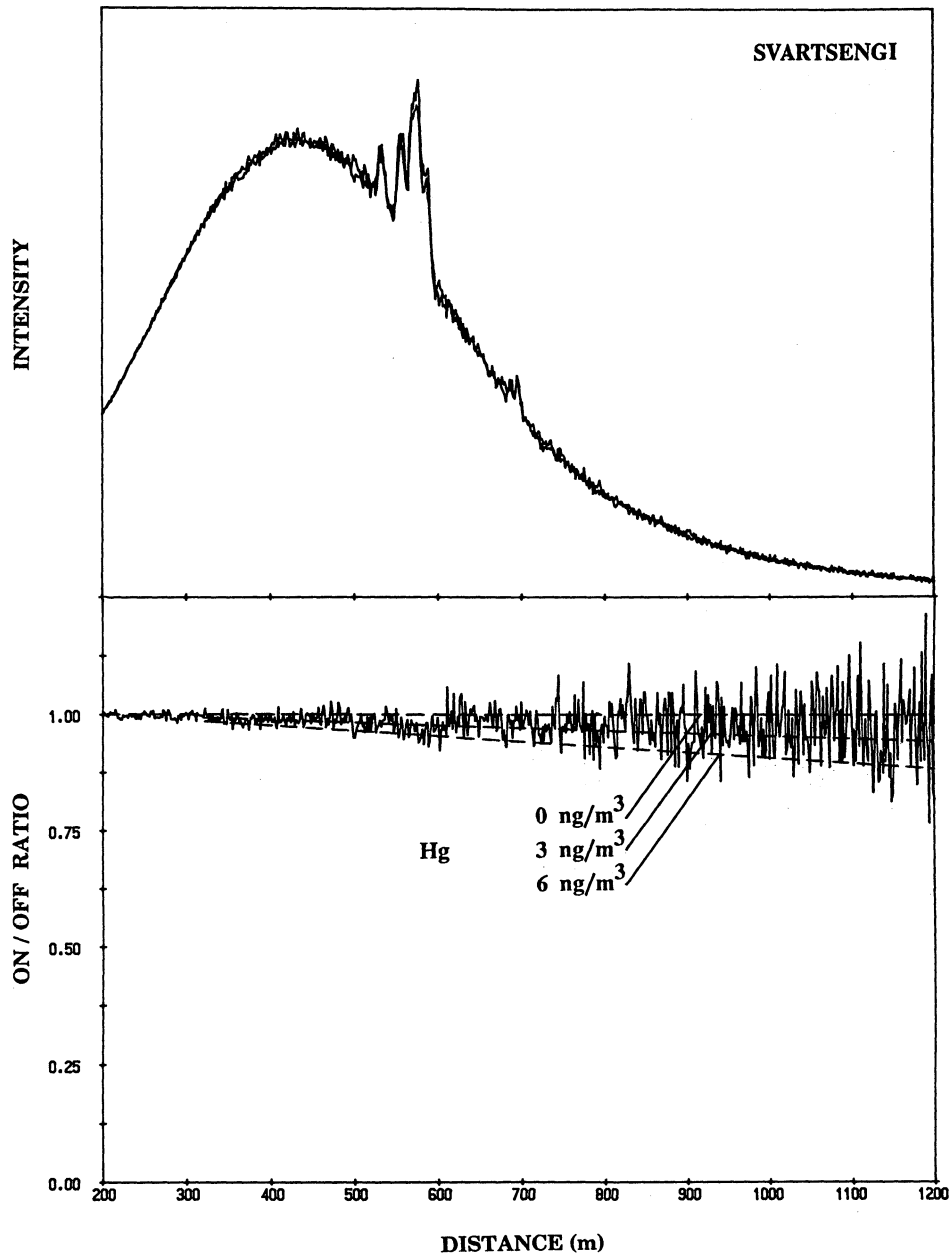


Fig. 4. On/off Hg lidar curves showing elastic plume returns and resulting DIAL curve. Theoretically calculated curves for uniform Hg concentrations of 0, 3 and 6 ng/m³ are included.

off-resonance lidar curves are shown superimposed on each other. For a homogeneous atmosphere the recorded backscattered intensity is expected to follow a $1/R^2$ dependence, as a function of distance R , for the case of no absorption. The observed curves, displaying a broad maximum at about 450 m range, are obtained because of the range-modulated photomultiplier gain switching. The gain is initially kept low but is ramped up to its full value, which is reached after about 3 μ s or 450 m. In this way the effective dynamical range can be extended for the receiver electronics. Between 500 and 600 m, structures due to the power-plant plume are encountered. At 700 m, further weak structures are observed. In order to detect any differences between the on- and off-resonance curves they are divided for each range value. In the lower part of Figure 4 the resulting divided (DIAL) curve is shown. If no differential absorption due to

the studied species is experienced the curves should be identical and the DIAL curve should have the value unity at all ranges. Clearly, the noise level will increase for greater ranges because of the lower backscattered intensities. On the other hand, the presence of a cloud of the gas will result in a falloff in the DIAL curve, from which the number densities at different ranges can be calculated, as has been done for the data presented in Figure 2. As is evident from Figure 4 no localized Hg clouds were encountered. In the figure, calculated DIAL curves corresponding to uniform background concentrations of 3 and 6 ng/m³ are included. By comparing these curves with the experimental one a background concentration of 1.5 (1.5) ng/m³ can be inferred.

Measurements were performed in many different directions for different weather and wind conditions. On one occasion a slightly elevated value was noted but not consis-

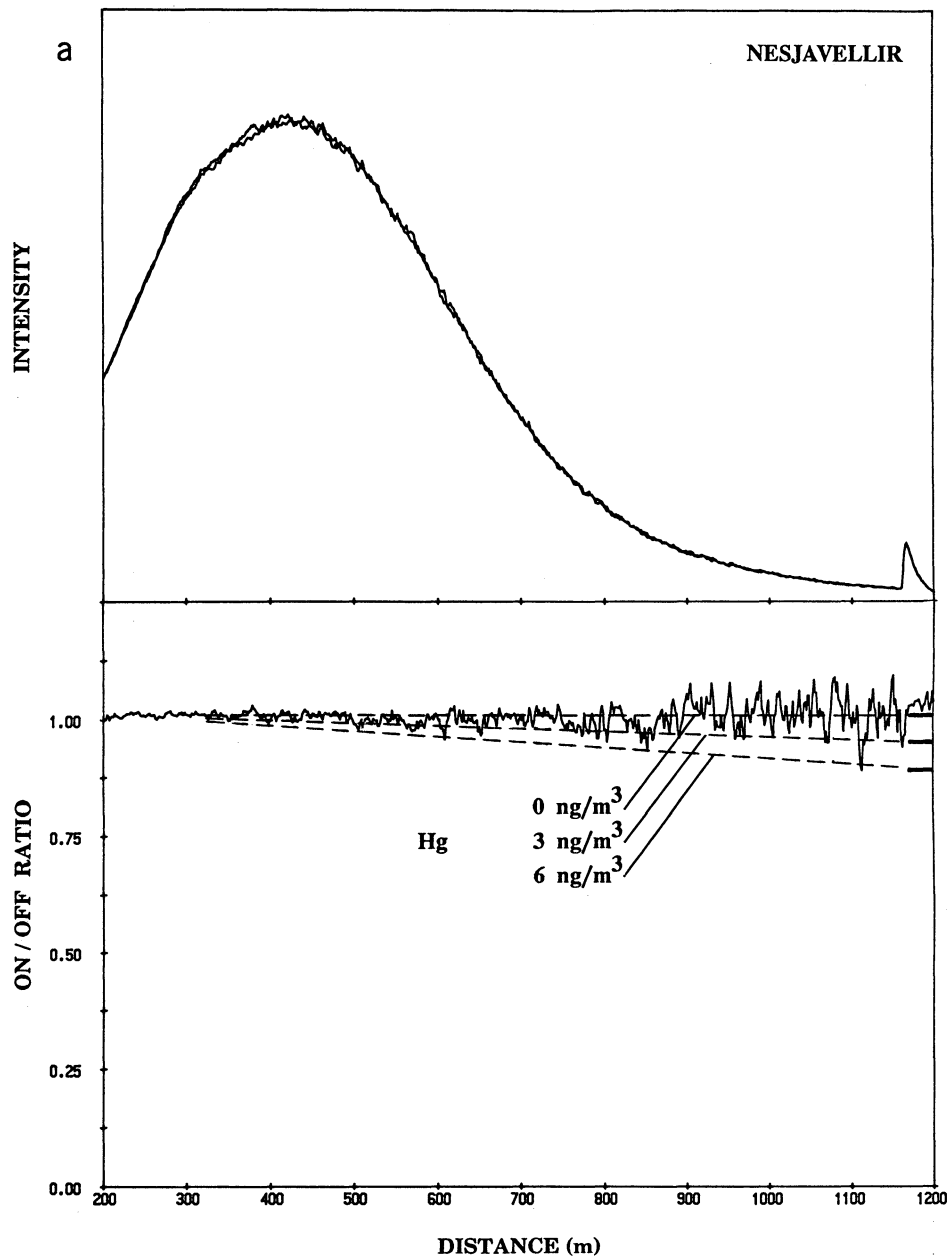


Fig. 5. (a) On/off lidar and DIAL curves for Hg. Theoretically calculated curves for uniform Hg concentrations of 0, 3, and 6 ng/m³ are included. (b) Corresponding curves for O₂ with concentration values for oxygen included. In both the Hg and the O₂ curves echoes due to a topographic target can be seen.

tently for a prolonged time. At one point, the dry steam well was vented to the atmosphere in an attempt to detect mercury by probing the resulting plume. We also performed measurements against retroreflectors placed in the vicinity of the Eldvörp seismic fault line traversing the area. However, no elevated concentrations could be recorded.

In order to try to shed light on these somewhat unexpected observations, concentrated well gas was collected and injected at the several percent concentration level in our White cell. The supporting vehicle carrying the White cell was placed next to the lidar system and the laser beam was transferred between the two buses for absorption measurements through the multipass cell. The cell measurements were also performed with time-resolved spectroscopy using signals derived from the beam entering and leaving the

multipass cell as data. No atomic mercury absorption line could be observed. Gas was also extracted and stored in Teflon bags for similar analysis in a stable laboratory setup in Lund with the same result. Assuming that no chemical reactions occurred between sample extraction and measurements on the same day in the field or a month later in the laboratory, an upper limit on the atomic Hg concentration, recalculated for concentrated Svartsengi well gas, is 0.25 $\mu\text{g}/\text{m}^3$.

Krysuvik

The Krysuvik geothermal field is located some 18 km to the east of the Svartsengi field. The upflow zones of the geothermal steam are below two major hyaloclastite ridges

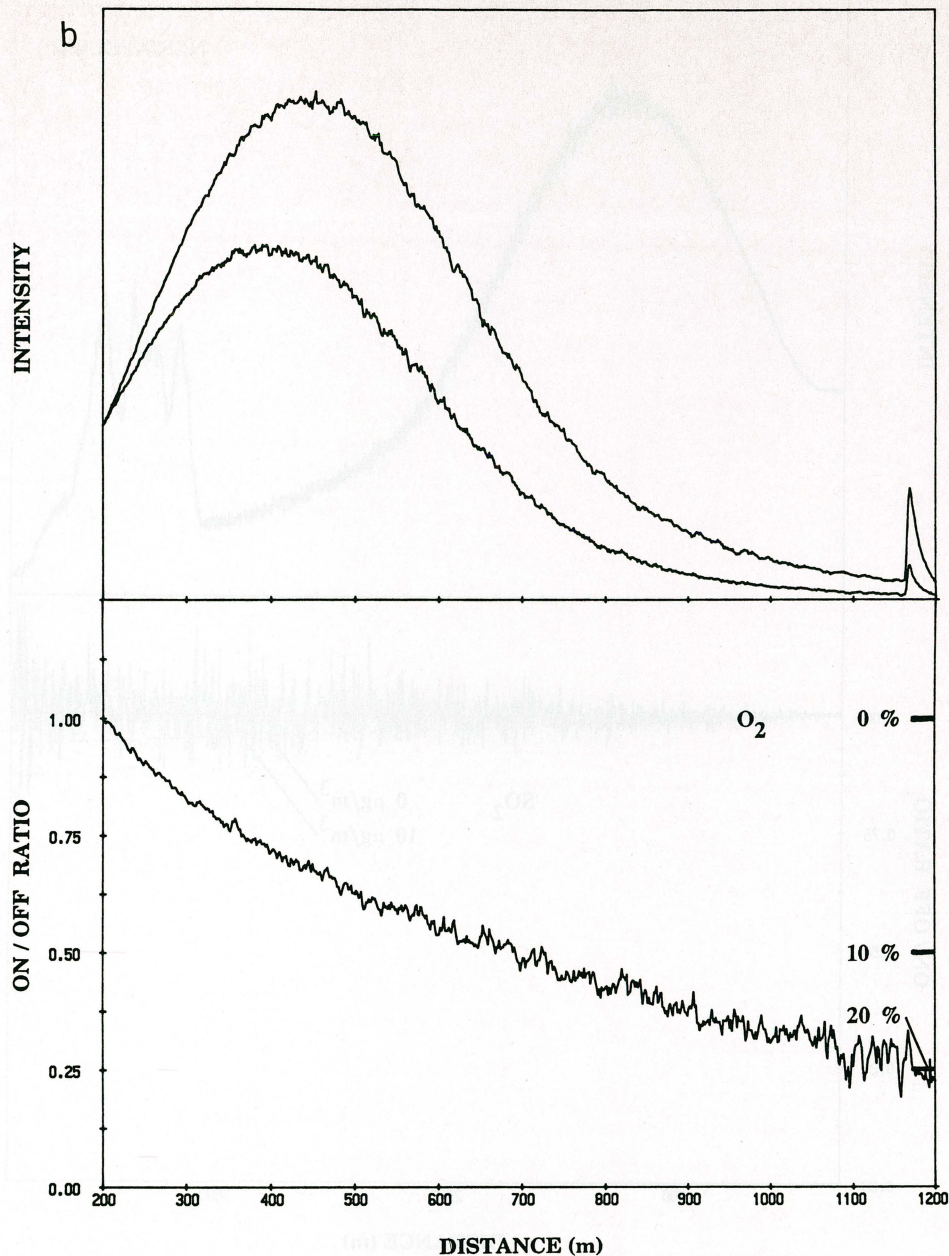


Fig. 5. (continued)

formed by subglacial eruptions during glaciation time. The field is intersected by two major fissure swarms. Reservoir temperatures, estimated from chemical geothermometers, are 260–300°C. Krysuvik is a largely undeveloped geothermal area. In contrast to the Svartsengi field, where the geogases are released from greater depths through the wells and energy extraction arrangements into the atmosphere, the Krysuvik area contains many surface manifestations with very active fumaroles and mud pots. Thus chemical reaction pathways might be expected to be different here. However, lidar measurements revealed no localized Hg plumes, and the maximum ambient air concentration of Hg, derived from the DIAL curve overall slope, was 1.5 (1.5) ng/m³. Flameless atomic absorption measurements of fumarolic steam samples yielded concentrations of ≥ 400 ng/kg, however.

Nesjavellir

Nesjavellir is a high-temperature geothermal area within the Hengill central volcano. The field is within a fault swarm north of Hengill, and many faults and volcanic fissures are observed at the surface. The valley is partly covered by a 2000-year-old lava, which is the youngest eruptive phase in the area. Reservoir temperature exceeds 320°C in the Nesjavellir field. This field is currently under development, and some 18 wells, with depths ranging to 2 km, have been drilled. The concentration of mercury in steam from the wells ranges from 300 to 1700 ng/kg, as determined by point measurements by the flameless atomic absorption method. Several of the wells were being vented into the open air through silencers during the test period. There are also

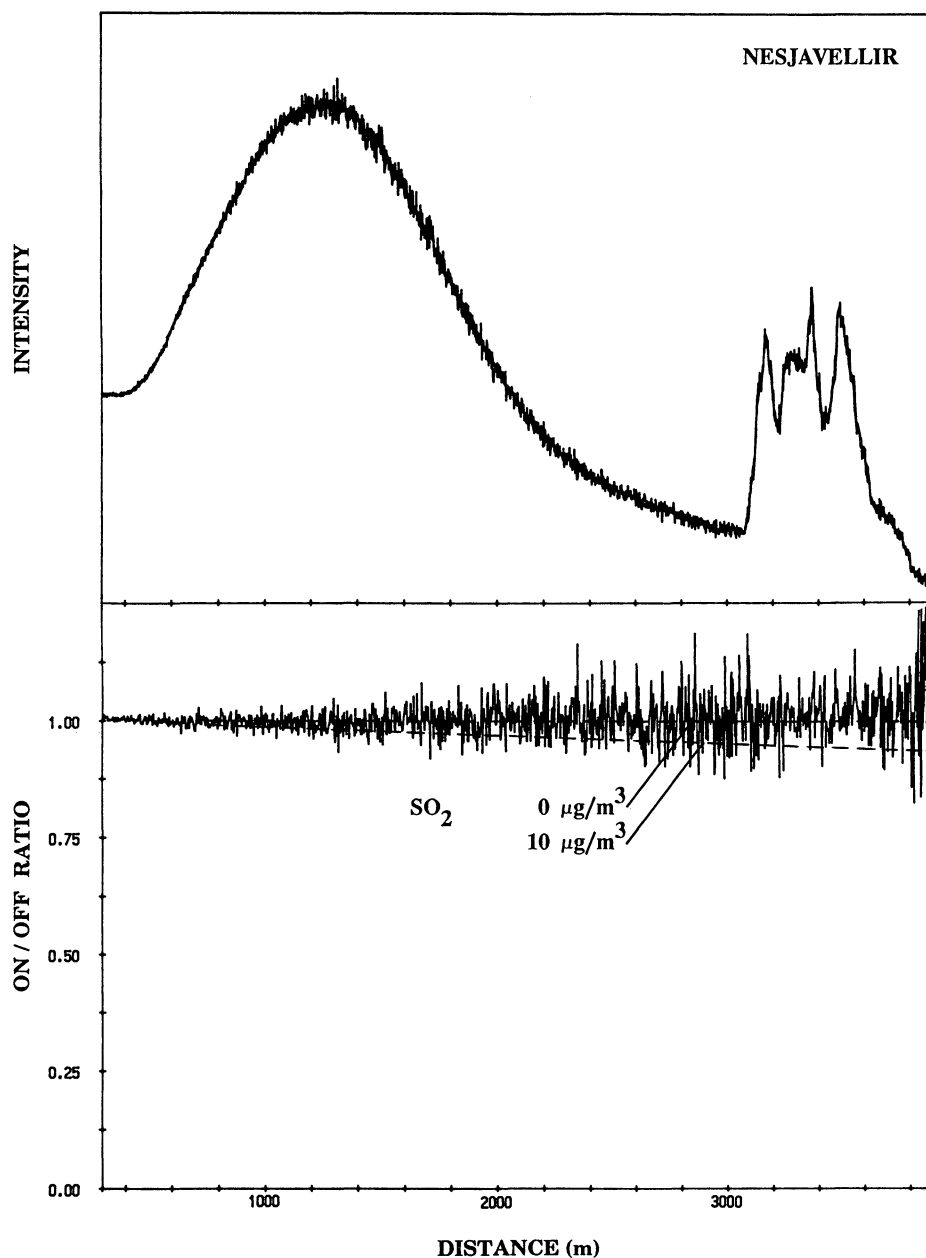


Fig. 6. On/off lidar and DIAL curves for SO_2 . Theoretical curves for 0 and $10 \mu\text{g}/\text{m}^3$ uniform SO_2 concentrations are included.

numerous active fumaroles and mud pots in this field. Measurements were performed in many different directions with regard to the well locations and the wind direction. A typical recording at Nesjavellir is shown in Figure 5a. In the upper part of the figure the on- and off-resonance lidar curves are superimposed, also displaying at 1200 m the optical echo from one of the hillsides surrounding the Nesjavellir valley. The input signal to the transient digitizer was here given a larger time constant in order to get better statistics of the otherwise very sharp echo. In the divided (DIAL) curve the topographic target is observed as a curve segment of smaller noise level due to the stronger localized signals. The hillside optical echo serves as a distant "optical power meter" for the transmitted laser beam and the ratio of the "readings" on and off the mercury line directly provides a number for the path mean concentration curves included in

the figure, no significant Hg concentration can be inferred, however, neither from the echo signal, nor from the general DIAL curve slope.

In view of the low concentrations recorded throughout the Icelandic field test, repeated verification of the validity of the measurement procedures used was sought. As already discussed, one obvious procedure was to accurately control the laser tuning to the mercury absorption line. A further system verification is illustrated in Figure 5b, displaying DIAL measurements of the uniform atmospheric O_2 concentration within minutes of the Hg test shown in Figure 5a. In the oxygen measurements an isolated weak Herzberg-band line situated a fraction of an angstrom from the Hg line was used. This line has a similar half width as the atomic mercury line. As can be seen, the presence of the natural oxygen is strongly manifested in the DIAL curve, providing an unam-

biguous check of the overall perfect functioning of the lidar system.

At Nesjavellir another aspect of geogases entering the atmosphere was investigated. The well gas contains high concentrations of H₂S. The question of the rate at which H₂S oxidizes to SO₂ downwind in the plume has remained an open one. As previously documented [e.g., Fredriksson *et al.*, 1981; Egebäck *et al.*, 1984; Edner *et al.*, 1987] the DIAL technique is extremely powerful in mapping atmospheric SO₂ distributions. With the very powerful laser system used in the Icelandic field test more than 10 mJ could be generated at the SO₂ DIAL wavelengths close to 300 nm. An example of data from Nesjavellir is shown in Figure 6, displaying on- and off-resonance lidar curves with distributed echoes from thin clouds at ranges between 3 and 4 km. Even after the cloud the atmospheric backscattering still remains. Actually, in the absence of clouds an effective range of about 6 km was reached. It was found that the atmosphere in the geothermal field was almost free from detectable SO₂. No concentration values above the noise limit, which corresponds to about 5 µg/m³ were detected, and no tendency of gradual downwind increase could be verified.

Clearly, our lidar investigations were aimed at producing total flux numbers for Hg and SO₂ from the geothermal areas using measured concentration values and wind velocities. However, since no concentration values elevated over the background values were obtained no net flux was detected.

DISCUSSION

The absence of elevated concentrations of atomic mercury in the air over at least three geothermal fields in Iceland presents a puzzle.

There can, on the one hand, be no doubt about the presence of significant amounts of mercury in well fluids in these areas. In the Svartsengi field, in particular, the total rate of transport of mercury to the surface is of the order of 250,000 ng/s. The concentration in the pond, to which all the brine and condensate are eventually discharged, is, on the other hand, only about 40 ng/kg, which is less than a tenth of the concentration in the well fluids. Furthermore, there are no obvious sinks in the power plant, such as cooling tower sludge, that might remove mercury from the fluids. Consequently, there is every reason to believe that most of the mercury is released to the atmosphere.

If a significant portion of the mercury were atomic, it should have been detected by the lidar search described above. Studies of geothermal areas elsewhere have, indeed, revealed that the greater part of the mercury released from these areas to the atmosphere is present as the element [Robertson *et al.*, 1977]. Thus, we would have expected to find mercury with the lidar technique, in concentrations well above the background.

The lidar technique is sensitive only to atomic mercury, however. Mercury in a form other than elemental vapor would therefore not have been revealed in this search. In this context, it is worth noting that Robertson *et al.* [1977] found that a lesser, but still significant, portion of the volatile mercury was present in forms that they listed collectively as Hg²⁺. At present, we thus consider the occurrence of the mercury in some form other than elemental vapor to be the most probable explanation for the failure of the lidar search to detect significant concentrations.

The question may be raised of whether rapid dispersal and dilution of the mercury in air is the reason for the low concentrations observed. Even if this were true for the mercury vented from the power plant and for that discharged from the open dry-steam well, it could clearly not be the case for the concentrated well gas collected into a bag. Dispersal does therefore not suffice as the explanation for the low concentration of atomic mercury.

Further research is planned to elucidate the fate of the mercury from the Svartsengi well fluids. It would be of particular interest to determine its oxidation state directly and to gain any other information about its chemical speciation in the area.

The geophysical conditions in Italian geothermal areas are different from those in Iceland. Lidar measurements in Italian geothermal fields and mercury mining areas are planned.

As a side result from the Icelandic campaign, it was found that the oxidation of H₂S in air at Nesjavellir was not rapid enough to produce elevated concentrations of SO₂, in spite of the continuous release of significant amounts of H₂S to the atmosphere.

The conclusion from the present work is that both atomic Hg and SO₂ are present in much lower concentrations in air over Icelandic geothermal fields than in Scandinavian or Western European urban or industrialized areas.

Acknowledgments. The authors are grateful to Nils Robert Nilsson, NORDITA, and Axel Björnsson, Orkustofnun, for support and encouragement in this Inter-Nordic Project. Valuable help provided by G. Sverrisdóttir and B. Galle is gratefully acknowledged. This work was supported by the Nordic Industrial Foundation and the Swedish Natural Science Research Council (NFR). One of us (G.W.F.) was supported by a grant from the U.S. National Science Foundation.

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J. Ö. Bjarnason, H. Kristmannsdóttir, and K. H. Sigurdsson, Orkustofnun, Geothermal Division, Grensásvegur 9, 108 Reykjavik, Iceland.

H. Edner, G. W. Faris, A. Sunesson, and S. Svanberg, Department of Physics, Lund Institute of Technology, Post Office Box 118, S-221 00 Lund, Sweden.

(Received January 9, 1990;
revised October 17, 1990;
accepted October 17, 1990.)