

ENVIRONMENTAL EXPOSURE TO HYDROGEN SULFIDE IN CENTRAL SLOVAKIA (RUŽOMBEROK AREA) IN CONTEXT OF HEALTH RISK ASSESSMENT

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SUMMARY

The activities of the kraft pulp-mill in Ružomberok have great impact on ambient air quality in the town and the neighboring villages. The malodorous sulfur compounds adversely contribute to the overall emission profile. The reduced sulfur proportion forms the inorganic and organic compounds containing sulfur atoms in their lowest oxidation condition (S^2). The total sulfur proportion reduced includes hydrogen sulfide, mercaptans, dimethyl sulfide, dimethyl disulfide and other sulfur compounds. Hydrogen sulfide (H_2S) – as mentioned above – is of prime importance. The kraft pulp-mill has measured of hydrogen sulfide contained in ambient air since 2002.

The environmental samples were collected in 7 localities in all their selection based on the geographic, climatic and demographic factors. Four exposure localities in the Ružomberok neighborhood, have been defined by the specified criteria. Exposure assessment was made for each exposure locality by determination of the average daily inhalation dose. The average concentration of H_2S reached $5.8 \mu g \cdot m^{-3}$ in the most polluted locality (Černová, 2003). To complete the health risk assessment, the standard risk characterization procedure was made by the Hazard Quotient (HQ) calculation for hydrogen sulfide exposure. The highest level of HQ, almost 7, was identified in the event of a worst case exposure scenario (using the 95% concentrations), as for exposure group C. Statistically significant decrease of reported H_2S levels was noted during the entire measurement period ($-0.25/\text{year}$, $p < 0.001$).

Key words: hydrogen sulfide, environmental exposure, Hazard Quotient, reference concentrations (RfC)

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INTRODUCTION

Ružomberok with its neighbourhood belongs to the most polluted areas in Slovakia. The local kraft pulp mill has exercised an influence on ambient air quality to a considerable extent, characterized by distinctive odor. Sodium sulfide in combination with sodium hydroxide used for the digestion of wood chips gives rise to this odor. The combined effect of sodium sulfide and sodium hydroxide enables delignification of wood within a relatively short period, while enabling the cellulose fibres to keep their strength. However, the incidence of sulfur gases such as hydrogen sulfide (H_2S), methyl mercaptan (CH_3SH), and methylsulfides ($(CH_3)_2S$ and $(CH_3)_2S_2$) (1), is deemed to be an unavoidable by-product of the mentioned process. Of the mentioned gasses, methyl mercaptan and hydrogen sulfide are considered to be predominant components. The presence of sulfur in a reduced state defines these compounds, collectively termed “Total reduced sulfur” (TRS), which have characteristic, specific odor at relatively low concentrations.

Hydrogen sulfide is a colorless, flammable gas with a characteristic of rotten eggs. The odor threshold is $0.0007\text{--}0.014 \text{ mg} \cdot \text{m}^{-3}$. Approximately 95% of H_2S present in the atmosphere is produced naturally from volcanic eruptions, sulfur springs

and biological decomposition (2). The primary anthropogenic sources of TRS include oil and gas processing facilities, kraft pulp mills, food processing plants, livestock feedlots and chemical manufacturing plants.

Human are exposed to exogenous hydrogen sulfide principally via inhalation and the gas is rapidly absorbed through the lungs. Penetration through the skin is negligible (3). The absorption rate in the gastrointestinal tract is not known. However, soluble sulfide salts present in the gastrointestinal tract (4) are capable of causing hydrogen sulfide poisoning.

Hydrogen sulfide is metabolized through three pathways: oxidation, methylation and reactions with metalloproteins or disulfide-containing proteins. Oxidation in the liver represents the major detoxication pathway. Thiosulfate is the major oxidation product, which is then converted to sulfate and excreted in urine. The methylation pathway also serves as a detoxification route.

Hydrogen sulfide is excreted primarily as sulfide in urine. It is also excreted unchanged in exhaled air and in faeces.

During oral administration of S-sulfide to rats, 50% of the dose of sulfate was found in urine after 24 hours (4). In case of parenteral intake 91% of the dose was found in urine (74%) and faeces (17%). Thiosulfate in urine is a useful indicator of hydrogen sulfide exposure (5).

Hydrogen sulfide toxicity results from its reaction with metalloenzymes. In the mitochondria, cytochrome oxidase, the final enzyme in the respiratory chain, is inhibited by hydrogen sulfide; this disrupts the electron transport chain and impairs oxidative metabolism. Nervous and cardiac tissues, which have the highest oxygen demand, are especially sensitive to the disruption of oxidative metabolism.

Most human data are derived from acute poisoning case reports, occupational exposure, and limited community studies. Single inhalation exposure to high concentrations of hydrogen sulfide causes respiratory, ocular, neurological, cardiovascular, metabolic and reproductive health effects. The respiratory, neurological, and ocular effects are the most sensitive end – points in humans following inhalation exposure. There are no adequate data on the carcinogenicity of H₂S.

At concentrations of 5 mg.m⁻³ and above hydrogen sulfide causes increased eye complaints, while at 15 mg.m⁻³ and above it causes conjunctival irritation (4). Serious eye damage is caused by a concentration of 70 mg.m⁻³. At higher concentrations (above 225 mg.m⁻³), hydrogen sulfide has a paralytical effect on the olfactory perception (4).

At higher concentrations, respiratory irritation is the predominant symptom. In healthy men and women the inhalation of hydrogen sulfide at 2.8–14 mg.m⁻³ for 16–30 minutes does not affect pulmonary function (8). At the concentration of around 400 mg.m⁻³, there is a risk of pulmonary oedema (4). Single exposure to >700 mg.m⁻³ hydrogen sulfide is deemed to be a reason for rapid respiratory failure (6).

In vulnerable groups – persons with asthma – bronchial constriction symptoms were detected at concentrations of 2.8 mg hydrogen sulfide mg.m⁻³ for 30 minutes (7).

Available information on the neurotoxic effects of single exposures to high concentrations of hydrogen sulfide in humans comes primarily from case reports. In most instances, exposure concentrations were either unknown or estimated. Single exposure to high concentrations of hydrogen sulfide can cause nausea, headaches, delirium, disturbed equilibrium, poor memory, neurobehavioral changes, olfactory paralysis, loss of consciousness, tremor and convulsions. At concentrations greater than 140 mg.m⁻³, olfactory paralysis occurs, causing a loss of odor perception; this makes hydrogen sulfide very dangerous, because a few breaths at 700 mg.m⁻³ can be fatal.

Epidemiological data concerning long-term exposures are limited. Hydrogen sulfide causes odor nuisance at concentrations far below those that cause health hazards. Typical symptoms and signs of hydrogen sulfide intoxication are most often caused by relatively high concentrations in occupational exposures. Low-level concentrations can occur more or less continuously in certain industries, such as in viscose rayon and pulp production, at oil refineries and in geothermal energy installations. Workers exposed to hydrogen sulfide concentrations of less than 30 mg.m⁻³ are reported to have rather diffuse neurotoxic and mental symptoms and in this regard have no statistically significant differences in comparison with a control group (9).

A recent-follow-up study provided further evidence that long-term exposure to low levels of malodorous sulfur compounds increases the risk of acute respiratory infection and pathological of respiratory tract symptoms (10).

The WHO air quality guideline for hydrogen sulfide determines 150 µg.m⁻³ was an average permissible limit concentration over 24 hours. As a result, the health end-point was eye irritation. To avoid odor annoyance, it is recommended that a 30-minute average ambient air concentration must not exceed 7 µg.m⁻³ (11).

MATERIAL AND METHODS

Study Design

The presented study has been implemented as part of the special project “The organic sulfur compounds impact on population health in Ružomberok neighborhood as a by-product from kraft pulp” (13). The project started in May, 2001, was carried out in 3 phases for 44 months and was supported by the Ministry of Health of the Slovak Republic. The study included 1) the standard design of the health risk assessment phases; 2) the effects assessment (hazard identification and dose – response assessment); 3) the exposure assessment and risk characterization. Afterwards, the risk management structure was identified under specific conditions. The Regional Authority of Public Health, Banská Bystrica, (Slovakia), acted as an expert sponsor of the project.

The following activities have been executed during the individual phases of the Health Risk Assessment:

1. Effects assessment – the available electronic and printed data sources were applied during this phase of work. This helped gather information of the physical-chemical properties, toxicity, acute, sub acute, sub chronic and chronic effects of the involved sulfur substances. Quality information assessment was also performed.

2. Exposure assessment – this phase focused on selection of the homogenous exposure areas within the territory in question, as well as the definition of the inhalation exposure levels within these areas.

3. Risk characterization – the quantitative risk characterization of hydrogen sulfide exposure was provided based on the Hazard Quotient calculation. The Hazard Quotient means the ratio of potential exposure to the reference concentration of a specific pollutant. In the event that the resulting Hazard Quotient is less than or equal to 1, there is no current assumption that adverse health effects will result from exposure. If the Hazard Quotient calculation exceeds 1, adverse health effects are likely to occur.

The Study Areas and Sampling Localities

The defined exposure groups of inhabitants were subjected to environmental exposure assessment in Ružomberok and neighboring villages. Selection of exposure groups, as shown in Table 1, was based on the following criteria:

- a) geographic and demographic factors (character of landscape, population density, etc.),
- b) climate factors, especially dominant wind directions,
- c) outputs of the concentration dispersion study, in which the paper mill factory has been identified as a main emissions producer,
- d) points of measurement in the locality.

The exposure group, with reference data of inhalation exposure, is shown in column 1 of the Table, whereby the measurement stations are listed in column 2. Columns 3 and 4 of the Table

Table 1. Distribution of exposed inhabitants by category of exposure and site of measurement

Exposure group/Total No. of people	Measurement station/code	Town/village	No. of participants
A/2178	Ivachnová (7)	Ivachnová	475
		Liptovská Teplá	885
		Bešeňová	398
		Liptovský Michal	261
		Madočany	159
B/2546	Martinček (4) Lisková (5)	Martinček	425
		Lisková	2121
C/8207	Černová (1a) Hrboltová (1b)	Černová	1430
		Hrboltová	676
		Rybárpole	1516
		settlement Kľačno	4585
E/2336	Štiavnička (6)	Štiavnička	539
		Liptovská Štiavnica	847
		Ludrová	950
Background	Liptovská Lúžna	Liptovská Lúžna	2940

include the villages and individual districts of Ružomberok with their specific population.

The criteria for localization of monitoring station were applied in accordance with recommendations the American Conference of Governmental Industrial Public Health Officers – selection for residential population exposure station. The station is to be located in the midst of a residential or suburban area but not in a central business district and must not be situated less than 100 m from any street with traffic volume in excess of 500 vehicles/day. Station probe height must be from 2.5 to 3.5 m. (12).

Samples (24-hour) were collected by continuous sampling for three years (2002, 2003 and 2004) during two periods each year – heating and non heating season.

MEASUREMENT METHODS

Different sampling techniques are available for measuring H₂S. Two general measuring classes have been developed. One of them is used for measuring industrial stationary source emissions and the other is based on ambient air quality monitoring. Continuous ambient monitoring survey together with direct reading analyzer is considered a method of choice for ambient air analysis, because of its ability to resolve short-term, infrequent episodes of high pollution which often represent a primary cause of concern (12).

In this case, the automatic gauging stations located in 9 places measured the concentration of H₂S in ambient air.

A mobile automatic gauging station was deployed in monthly intervals, alternatively in Ivachnová, Martinček, Lisková, Černová, Hrboltová, Štiavnička and Liptovská Lúžna (background) localities.

Pulsed fluorescence technology was applied for continuous measuring of H₂S. Initially, all SO₂ in the air sample was scrubbed out so that it did not interfere with the measured H₂S concentration. The remaining H₂S content in the sample was then converted to SO₂. Afterwards, the sample was drawn through a

sample chamber where it was irradiated with pulses of ultra-violet light. Any SO₂ in the sample was excited to a higher energy level and upon returning to its original state, light or fluorescence was released. The amount of fluorescence measured was proportional to the SO₂ (converted from H₂S) concentration.

Data Analysis and Interpretation

Standard methods were applied in the data analyses. The basic statistical characteristics for each measurement station (arithmetic mean, maximum and minimum value, 75 and 95 percentile), necessary for exposure definition, were calculated using MS Excel software. The pollutant concentrations at 95 percentile have been used for the worst exposure scenario and those at 75 percentile for the average emission scenario in accordance with the standard approach. The overall exposure concentration of total reduced sulfur and hydrogen sulfide in case of individual exposure groups A, B, C, E calculating arithmetic means and 75 and 95 percentiles per each year, measured for each exposure category in the relevant points of measurement.

RESULTS

Hydrogen Sulfide Concentrations

Table 2 shows the general results of H₂S measurements. In 2002 measurements using mobile stations started at five measurement sites, supplemented with the additional four monitoring points in 2005 (including the background station).

The hydrogen sulfide sulfur concentrations have been measured since 2002 at 7 measurement sites. The average levels varied from the locality to locality. During 2002 and 2003, the highest concentrations were detected in the Černová point of measurement, and in the Martinček point of measurement since 2004. The

Table 2. Measurements of hydrogen sulfide on individual measurement sites

Locality	year	min ($\mu\text{g.m}^{-3}$)	mean ($\mu\text{g.m}^{-3}$)	75 perc. ($\mu\text{g.m}^{-3}$)	95 perc. ($\mu\text{g.m}^{-3}$)	max. ($\mu\text{g.m}^{-3}$)	Samp. No
Černová	2002	1.0	3.4	4.4	6.1	9.8	61
	2003	1.1	5.8	7.0	14.1	21.1	56
	2004	0.8	1.6	1.9	2.1	2.5	59
Ivachnová	2002	0.4	1.2	1.5	1.8	1.8	30
	2003	0.2	1.2	1.4	1.9	2.4	60
	2004	0.7	1.4	1.6	1.9	3.6	42
	2005	0.2	0.8	0.9	1.3	2.4	72
Lisková	2002	0.1	0.9	1.3	1.9	2.6	30
	2003	0.6	2.1	2.7	3.6	5.1	90
	2004	1.2	1.9	2.2	2.6	2.8	29
Martinček	2002	0.9	1.6	1.8	2.3	2.4	30
	2003	0.4	1.9	2.8	3.4	5.7	59
	2004	0.3	2.5	3.0	5.8	8.2	89
	2005	1.4	2.1	2.3	4.4	5.1	90
Štiavnička	2002	0.0	0.5	0.7	1.1	2.3	31
	2003	0.0	0.6	0.9	1.3	1.6	57
	2004	0.0	1.1	1.4	2.0	2.5	90
	2005	0.0	1.6	1.9	4.3	4.8	71
Hrboltová	2004	1.5	2.1	3.2	3.8	4.2	31
	2005	0.2	1.6	2.1	3.5	4.0	66
Background	2005	0.2	1.7	3.6	3.7	4.0	19

lowest values measured during the entire measurement period were recorded at the Štiavnička measurement point (Fig. 1).

These measurement results were necessary to specify the exposure concentrations detected in the individual localities of Ružomberok neighborhood. Two separate exposure scenarios were prepared – the worst scenario representing the 95 percentile values and the average exposure scenario of the concentration values at 75 percentile. Table 3 and 4 show the calculated exposure concentrations.

There are the striking exposure differences between the worst and average exposure scenarios. The aim of the worst scenario modeling is to deal with situations that are unlikely to happen in practice; however, this may happen under the specific conditions. In general, the average exposure scenario deals with the usual exposure situations regarding preservation of sensitive population groups (children, elderly people, etc.).

The distinctions between the individual exposure groups underwent tests using the standard linear regression analysis. Concerning the hydrogen sulfide exposure levels, there was a significant difference defined in the two exposure groups – A (+0.97, $p=0.012$) and C (+0.93, $p=0.016$).

A significant statistical decrease was reported for H_2S during the entire measurement period ($-0.25/\text{year}$, $p<0.001$).

Hazard Quotient Calculation

The Hazard Quotient was expressed for both the worst and average exposure scenarios as a ratio of the exposure concentrations in the individual exposure groups and RfC (0.002 mg.m^{-3}).

Thus, the Hazard Quotient (HQ) is relevant for the average daily exposure levels. The following Table 4 shows the HQ values in single exposure groups in both exposure scenario cases.

While the Hazard Quotient exceeds the value in Exposure group C on a regular basis throughout the measurement, the Hazard Quotient for exposure group B varies at level 1.

However, the worst case scenario presents a for more adverse situation. The Hazard Quotient exceeded 1 in exposure groups B and C during the whole period, and in group E it exceeded 1 in 2005. There are much higher Hazard Quotient values than in the case of the average exposure scenario.

DISCUSSION

The present report represents the first study analyzing the results of malodorous sulfur measurements in ambient air in Slovakia. Its results are believed to be important for the environmental health sector. The differences in the ambient air concentrations of H_2S which occur in the areas where the kraft pulp-mills are located, are dealt with in many literature sources. Canadian sources can provide valuable and high quality data in this respect.

Hydrogen Sulfide Concentrations

The results from Ružomberok are comparable to with a large study implemented in Canada (Alberta). The outputs from both

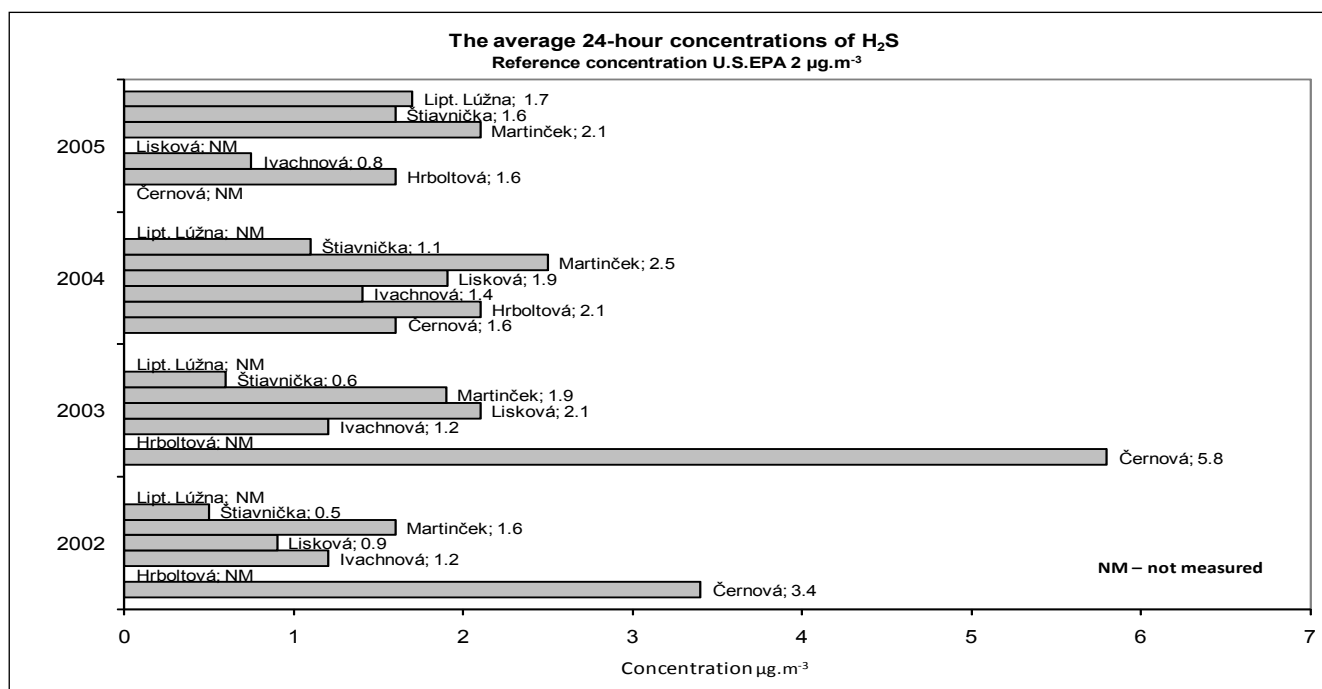


Fig. 1. Average 24-hour concentrations of hydrogen sulfide in single measurement sites.

Table 3. Exposure concentrations for the worst and average case exposure scenario

Exposure group	Relevant stations ^a	Statistical function	H ₂ S (µg.m ⁻³)
Worst case exposure scenario			
A	7	mean calculated using 95 perc. in 2002–2005	1.7
B	4, 5	mean calculated using 95 perc. in 2002–2005	3.4
C	1a, 1b	mean calculated using 95 perc. in 2002–2005	5.9
E	6	mean calculated using 95 perc. in 2002–2005	2.2
Background	8	95 percentile in the year 2005	3.7
Average case exposure scenario			
A	7	mean calculated using 75 perc. in 2002–2005	1.4
B	4, 5	mean calculated using 75 perc. in 2002–2005	2.3
C	1a, 1b	mean calculated using 75 perc. in 2002–2005	3.7
E	6	mean calculated using 75 perc. in 2002–2005	1.2
Background	8	75 percentile in the year 2005	3.6

^a The stations codes see in Table 1

of these measurements do not reveal a great deal of variance. However, such a comparison is not absolutely correct because the sampling methodologies are quite different. We have done only purposive sampling, especially for economic reasons. Ideally, we should have a representative number of samples which would be much better suited to such a exposure assessment. This has to be taken into consideration when interpreting the outputs.

Eighty monitoring stations operating in Alberta analyzed the H₂S ambient concentrations for three years. The stations are located in areas affected by oil and gas exploration and production facilities such as sour gas wells, gas and crude oil batteries, gas and oil processing plants, oil refineries, and tar sands mining and

processing. Alberta Environment (AENV) monthly results for the period from January 1999 to November 2001 included 1,653 entries of the H₂S maximum concentrations (1-hour averaging time). If the three highest concentrations were excluded, the average concentration of H₂S (1-hour highest) from anthropogenic sources based on three years of monitoring reached 0.00602 ppm (6.02 ppb or 8.5 µg.m⁻³) in Alberta (2).

Health Effects

Jaakkola et al. reported that exposure to hydrogen sulfide in people living in the area with the pulp-mill caused 12 times

Table 4. The Hazard Quotient values

Exp.group	A	B	C	E
Worst case exposure scenario				
2002	0,9	1,1	3,1	0,6
2003	1,0	1,8	7,1	0,7
2004	1,0	2,1	1,5	1,0
2005	0,7	2,2	1,8	2,2
Average exposure scenario				
2002	0,8	0,8	2,2	0,4
2003	0,7	1,4	3,5	0,5
2004	0,8	1,3	1,3	0,7
2005	0,5	1,2	1,1	1,0

more eye irritation compared to people not exposed to hydrogen sulfide (7). These effects were observed at the mean annual hydrogen sulfide concentration of $6 \mu\text{g}\cdot\text{m}^{-3}$. However, the reported ocular symptoms may have been caused by the exposure to peak concentrations of hydrogen sulfide (daily peaks as high as $100 \mu\text{g}\cdot\text{m}^{-3}$) or co-exposure to methyl mercaptan and methyl sulfides. A similar situation is very likely to occur in the Ružomberok area as well under certain combined circumstances.

Series of studies reported the results of the South Karelia Air Pollution Study which, in 1986, began to evaluate the effects of a low-level mixture of air pollutants on human health from the pulp-mills in South Karelia and Finland (10). The pollutant mixture included particles, sulfur dioxide and a number of malodorous compounds, including hydrogen sulfide, methyl mercaptan, and methyl sulfides. In the early studies of this series the levels of hydrogen sulfide, sulfur dioxide, particulates and methyl mercaptan were individually reported. In the following studies, a complex mixture of malodorous sulfur components was monitored in the form of total reduced sulfur (TRS) using a method that first removes any sulfur dioxide, then oxidizes the TRS compounds to sulfur dioxide and reports the results as micrograms per cubic meter. The information provided does not enable identification of the hydrogen sulfide TRS proportion, although the authors indicate that it is about two-thirds (7). These studies have demonstrated that low levels of hydrogen sulfide in combination with other sulfur-containing pollutants and, possibly, in combination with particulates and/or sulfur dioxide can have an adverse effect on respiratory health. However, at present we cannot determine whether this means the low annual average TRS levels at $1.2 \mu\text{g}\cdot\text{m}^{-3}$ or the daily average concentrations $56 \mu\text{g}\cdot\text{m}^{-3}$, which are associated with these findings.

The levels measured in Ružomberok neighborhood foreshadow quite similar results. The health effects can possibly be assumed based on the preliminary results.

CONCLUSIONS AND RECOMMENDATIONS

It is necessary to implement systematic epidemiological research concerning relevant health effects within this region. The results of this study will form the basis for appraisal of health risk management effectiveness. Health management recommendations have been formulated, focused on the monitoring of ambient air quality steps and public information strategy. The health risk communication has been identified as one of the priority areas.

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