THE IMPACT OF METEOROLOGICAL PARAMETERS ON SULPHURIC AIR POLLUTION IN KOHTLA-JÄRVE

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The hydrogen sulphide (H_2S) and sulphur dioxide (SO_2) data from Kalevi air quality monitoring station in the town of Kohtla-Järve, the centre of Estonian oil shale processing industry, and some episodic measurements from a nearby location, are analysed in view of wind direction and other meteorological parameters governing the atmospheric dispersion. Unlike in a typical urban airshed, sulphur compounds in Kohtla-Järve constitute a major air quality problem, H_2S sometimes exceeding the permitted limit value. At Kalevi monitoring station two narrow wind direction intervals are identified as related to high concentrations. According to time-dependent pattern and wind direction analysis at different heights, these two peaks are most likely originating from the oil-shale retorting plant and the Kohtla-Järve municipal wastewater treatment plant. Due to active reconstructions of these plants, both concentration peaks have been decreased during recent years.

Introduction

Air quality concerns typical of contemporary urban environment are nitrogen oxides and volatile organic compounds in busy city centres and ozone as a secondary pollutant in outskirts [1]. In addition, higher carbon monoxide concentrations are common, but usually not reaching the maximal permitted limits. Sulphur dioxide has been an urgent air quality problem in many countries, nowadays often related with marine ship routes [2]. The exceptionally high (but within the limit values) hourly average SO₂ concentrations about 60–70 μ g/m³ are measured in Estonia, near the passenger port of Tallinn [3]. Remarkable quantities of H₂S are emitted also from fuel oil terminals [4], animal farms [5] and municipal waste treatment sites [6].

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Unlike most European urban municipalities, the Estonian oil shale processing center Kohtla-Järve has often to deal with enhanced concentrations of H_2S and SO_2 in the ambient air, H_2S sometimes exceeding the regulatory limit level 8 µg/m³. Three main enterprises known as remarkable emission sources of sulphuric compounds are governed by Viru Chemistry Group (VKG) – the oil-shale retorting plant of VKG Oil (shale oil production unit and extensive semi-coke dumps, identified as important sources of H_2S [7]), thermal power stations of VKG Energia (oil-shale and generator gas combustion are the significant sources of SO_2), and the municipal wastewater treatment (WT) plant that treats both residential and oil shale processing sulphur-rich wastewater. Sulphide formation in the 16 km long pressure pipe (ongoing wastewater from the industrial clients), where the wastewater retention time is 24 hours, has been investigated [8].

To date the shale oil production processes implemented in Estonia are the *Kiviter* process (particle size 25-125 mm) and the *Galoter* process (particle size < 25 mm) [9]. The new plant Petroter which is based on the Galoter process started to operate recently.

Production of oil from lumpy oil shale in vertical retort, the Kiviter process, results in large amounts of solid waste – semicoke. Per tonne of oil shale 0.49 tonnes of semicoke are formed in vertical retorts. Semicoke contains a considerable amount of sulphur (1.7–2.1%) in different forms [10], since in the retorting process of Estonian oil shale, more than 50% of sulphur in the raw oil shale remains in the solid residue afterwards [11]. Figure 1 illustrates the air emissions from the retorting process. The maximal emission quotes according to air pollution permission are indicated. Thus, the figures do not correspond exactly to any particular year discussed below, but to the upper limit for permitted emissions.

In the past as well as in present, besides shale oil, several chemical products have been produced: bitumen, coke, phenols, phenolic products, etc.

Besides emissions from controlled industrial processes, there are landfill emissions. The surface area of the semi-coke landfill is 142 ha, plus the area needed for treating the wastewater. Approximately 73 million tons of industrial waste has been deposited in this landfill at present. Semi-coke and oil shale ash are not the only two types of industrial waste being deposited in the landfills. However, a variety of the types of waste used to be wider – fuses (pitch waste) and acid tar (originating from the production processes of benzene and toluene), waste containing sulphur and arsenic (so-called sulphur sludge, originating from the refinement of the generator gases, where As₂O₃ sodium carbonate solution was used), waste containing mineral oil, building rubble, and consumer waste, originating from the activities of the enterprises, have been deposited in the industrial waste landfill. In addition, hazardous wastes from AS Velsicol and AS Nitrofert have been deposited in the industrial waste landfill over the years. The former landfill for industrial waste has also been used for depositing building and demolishing rubble



Fig. 1. Emissions of SO_2 and H_2S from technological processes of AS VKG Oil. The maximal emission quotes according to air pollution permission are indicated.

(including wood wastes), sediments of waste water, street sweepings, and probably also domestic waste [12].

There is a permanent air quality monitoring station at Kalevi street located a few kilometres down the dominating western and south-western winds from both VKG Oil and wastewater treatment plant (Fig. 2). Despite extensive measurements both in the town and at industrial territories, the origins of certain air pollution episodes still remain obscured. Narrow peaks of elevated concentrations with nearly south-western wind directions are mentioned through several years [13, 14] – roughly originating from the direction where the industrial enterprises are situated, but certain directions (azimuthal angles approximately 205–210° and 220–230°) do not correspond exactly to any of known pollution sources.

This paper aims to identify the origins of periodically high H_2S and SO_2 concentrations at Kalevi monitoring station. For that the coinciding wind directions at different locations and heights are analysed. The meteorological (weather forecast) modeling results are applied as a supplement to scarce local measurement data of vertical wind spread. The sonic anemometer measurement data are used to gain information about the impact of atmospheric boundary layer properties on the local pollutant transport. In addition,

it is tested to which extent the improvements of technological processes in VKG have reduced the levels of sulphuric pollutants in the air of the town of Kohtla-Järve.



Fig. 2. Locations of (a) monitoring stations and known pollution sources in the study area, base map by Estonian Land board (Maa-amet) and (b) monitoring stations in Ida-Viru county (North-East Estonia) and Aseri meteorological mast.

Data and methods

Air quality monitoring data originate from Kalevi monitoring station in Kohtla-Järve and mobile station Mobair operated by Estonian Environment Research on the territory of VKG Oil in November 2005 - January 2006. Meteorological data were gathered from monitoring stations, Aseri meteorological mast operated by Estonian Environment Research (see Fig. 2), and meteorological (weather forecast) modeling results. The meteorological modeling results, applied for 2005/2006 winter episode only to compensate the lack of measurements higher above the ground, originate from model ETB HIRLAM operated routinely by the Estonian Institute of Meteorology and Hydrology since January 2006 and BaltAn65+ reanalysis [15] (November - December 2005). The automated monitoring station in Kohtla-Järve is situated at Kalevi street (geographical coordinates 59°24'35" N and 27°16'43" E). This station meets the requirements of the European Environmental Agency and belongs to European monitoring net EUROAIRNET [16]. The measurements of pollutant concentrations are provided all year round, representing the hourly mean concentrations.

Mobair is a mobile unit for air monitoring that was installed in Kohtla-Järve near the main entrance to VKG AS production area during 29.11.2005-17.01.2006.

From air monitoring stations and Mobair only the low-level (about 3 m) wind speeds, directions, atmospheric pressure and temperature are available. Additionally there is a metrological mast in Aseri (located 29.5 km to northwest from Kalevi monitoring station) measuring temperatures at 8 and 20 m, wind speeds and directions at 10 m and 24 m levels and the vertical wind dispersions. The meteorological model data are pre-processed with the air quality model SILAM [17]. The data include meteorological parameters at several levels (for instance, 15, 55, 130, and 305 m): wind speeds and directions, turbulent diffusivities, etc.

The emissions according to Estonian national statistics are given in Fig. 3. Data is gathered from registers of air emissions and permitted emission quotes of waste-water treatment plants by Estonian Environment Information Centre (Keskkonnateabe Keskus), requested in August 2010. The sharply increased SO₂ emissions in 2005 are caused by the growth of shale oil production, conditioning the increased combustion of generator gas at thermal power plants. In the second half of 2008 a sulphur-catching device (NID-reactor) was installed at VKG Energia Põhja (Northern) Thermoelectric Power Plant (TPP), providing 65% effectiveness in extracting SO₂ from exhaust gas. Hydrogen sulphide emissions in 2007 from VKG Oil are *circa* 5 times less than in 2006, that is explained by reconstruction of the park of tanks and installation of collective breathing system with absorbers. Emissions of earlier years, up to 2006, from the wastewater treatment plant are obviously underestimated as unrealistic emissivities were applied. About five-fold correction was based on measurements by Estonian Environment

Research [14]. The higher emissions are applied for 2007, but earlier ones remained untouched in statistical database, although the real values should be close to 2007. In 2007 the regional wastewater treatment plant was reconstructed, and together with environmental measures by VKG Oil, that conditioned the decrease in emissions in 2008 (Fig. 3).



Fig. 3. Annual emissions of SO₂ and H₂S from main factories of Kohtla-Järve. Due to lack of data about Fortum Termest, operating the thermal power plants that time, the amount of 2003 was taken for SO₂ emissions in 2004. For Järve wastewater treatment (WT), 2007–2009, data about H₂S was taken corresponding to maximal quote according to air pollution permission.

Analysing the observations, the horizontal mass fluxes of H_2S and SO_2 ($\mu g/m^2 s^{-1}$) are used rather than concentrations ($\mu g/m^3$). The mass flux is defined as the mass of admixture transported through the unit surface in unit time, i.e. concentration multiplied by wind speed. As our goal is to understand the origins of pollution rather than to quantify the exceedings of permitted limit values, the fluxes are preferred, as they are stronger attributed to the pollution source strength (effect of downwind dilution is eliminated). The flux applied here is always calculated as the concentration multiplied by the wind speed measured in the monitoring station, even if the wind direction applied in the analysis originates from any higher level. Wind speed, of course, increases with height, but usually remains well correlated with low-level speed. Thus, we can still compare the fluxes at different time moments. On the other hand, as concentrations are not measured at higher

levels, we have no solid ground to apply the upper-level wind speeds for calculating the fluxes.

Results

Dependence of measured concentrations on wind direction is given in Figures 4 and 5. Automatic monitoring of H_2S concentrations at Kalevi station was started in 2004. H_2S fluxes have stably high values with wind directions between 195–250° (i.e. nearly southwest), but highest values are



a) H₂S 2006 - 2007

Fig. 4. Distribution of H_2S (a) and SO_2 (b) fluxes during 2006-2007 plotted against wind direction at Kalevi station (height 3 m) and Aseri meteorological mast (height 24 m). The fluxes are moving averages over 201 single measurements or nearly 4° of direction.



Fig. 5. Distribution of H_2S (a) and SO_2 (b) fluxes during 2008-2009 plotted against wind direction at Kalevi station (height 3 m) and Aseri meteorological mast (height 24 m). The fluxes are moving averages over 201 single measurements or nearly 4° of direction.

observed in a more narrow peak, $215-235^{\circ}$. In 2007 H₂S peak becomes narrow and low lumped in between $195-225^{\circ}$, remaining practically unchanged through 2007–2009. Sulphur dioxide peak ($195-225^{\circ}$) is clearly visible during 2005–2009 and has a nearly constant height, presumably indicating at a single source or compact group of sources. Thus, according to recent measurements, both the H₂S and SO₂ originate from one direction, $210 \pm 15^{\circ}$, whereas earlier observations (before reconstruction of wastewater treatment) give a stronger H₂S peak at $225 \pm 15^{\circ}$ without backgroundexceeding levels for SO₂ from the same direction. Assuming that 210° peak originates from VKG (Energy and Oil) and 225° peak from the wastewater treatment plant, it is easy to explain such a pattern, as the process in WT do not emit any remarkable amount of SO₂. But looking at the location map (Fig. 2), we recognise that these directions do not match with exact locations of VKG and WT: the directions from Kalevi monitoring stations are 215–230° and 240–250°, respectively. Thus, both directions are matching the edge of peak, leaving the highest values out of range. However, the discussed wind directions are measured at the low level in the monitoring stations under the direct impact of underlying surface elements, while the transport of air mass does not match exactly with low-level wind directions, as atmospheric air is typically mixed through much thicker layer than a few meters, when transported a few kilometres downwind. Thus, the higher-level winds are examined.

Comparison of fluxes plotted against the 3 m (Kalevi) and 24 m (Aseri) wind directions during 2006–2007 (Fig. 4) shows a systematic shift of peaks by 10–15° westwards. Remarkable is that despite slightly larger scatter, the general structure (including two peaks of H₂S) is preserved. The same is valid for 2008–2009 (Fig. 5), when only one (the southernmost) peak for H₂S is left. It is evident that these directions fit much better with positions of expected sources in respect to Kalevi station: 210–230° for VKG and 230–250° for the wastewater treatment plant. The fluxes presented at graphs are highly smoothed, using a moving average over wind directions. The single measurements often give values several times higher.

The direction-dependent (local wind direction in Kalevi) concentration distributions during the winter episode 29.11.2005–17.01.2006 are given in Fig. 6. Like during entire long-term monitoring series, the fluxes of SO₂ form a narrow peak around 205° and H₂S has more scattered high-level range, possibly with two peaks, at 195–240° (each data point presents an hourly value without any directional averaging, thus single maximum values are much higher than in Figures 4 and 5). The Aseri 24 m wind directions give expected Ekman shift of the peak by 13° clockwise with standard deviation 11°. Meteorological model results give systematically clockwise turning wind with height as well: by 1, 4, 9 and 23° for 15, 55, 130 and 305 meter levels, respectively. But their scatter is rather large: standard deviations ranging from 15 to 28°.

In Mobair station the high levels occur between 170 and 260° (south to south-west-west) with two sharp peaks at 170–180° and 225–240°. The production units of VKG are situated in south-south-western to western directions or 200–270° from this station (Fig. 2). As the buildings of VKG in these directions, immediately surrounding the Mobair station, are several times higher, severe and complicated distortions of wind field at this site are expected. Thus, it is quite evident that these highly elevated levels originate from production units of VKG Oil, the SO₂ sources are shale oil distillation and electrode coke production units, and possibly, from VKG Energia Lõuna (Southern) Thermoelectric Power Plant (TPP) as well. H₂S emissions from 200-270° probably originate from gas-generator plants and electrode-coke unit. Much lower peak of H₂S from north-western to northern directions may originate from the wastewater treatment plant.



Fig. 6. Fluxes of SO_2 and H_2S at Kalevi (a) and Mobair (b) monitoring stations, depending on local wind direction, 29.11.2005–17.01.2006.

Now we have fixed the directions, where the high levels of sulphurous pollutants come from. To examine further the atmospheric conditions that favor the highest levels, we leave out all wind directions as "noise" not containing information about local sources, except the range of $195-240^{\circ}$, and analyse the micrometeorological data from the Aseri mast in view of SO₂ and H₂S fluxes.

In Fig. 7 there are given the monthly average thermal stratifications at lower (8–2 m) and upper (24–8 m) layer at night and day (to include more statistics, wind direction segregation is not applied here; but 195–240° are proven similar in general). At night-time the potential temperature is increasing with height, indicating prevailing thermal inversions that are extremely

strong in summertime, when daily temperature range is larger. In the daytime in summer the unstable stratification (decreasing potential temperature) is prevailing due to solar heating in the lowermost layer, whereas the upper layer is rather neutrally stratified. All the daytime and night-time fluxes, especially of SO₂, are larger in winter and decrease in summer (Fig. 8). The gap for summertime night SO₂ is very deep – that may be conditioned by near ground inversions.



Fig. 7. Vertical potential temperature gradients (Kelvin per meter) at Aseri mast (2005-2010) at night-time (a) and daytime (b).



Fig. 8. Monthly average horizontal fluxes at Kalevi monitoring station (2006–2009). Only wind directions most favourable for local pollution sources ($195-240^{\circ}$) are included.

Correlations between Aseri vertical temperature gradients and sulphuric pollutant fluxes at Kalevi station are negative (*i.e.* inversion conditions tend to make less pollution transported at low air level), but small and insignificant. The vertical wind standard deviations correlate better with fluxes, especially for SO₂ (correlation coefficient +0.43 at 10 m level). The positive correlations indicate that stronger vertical mixing (*i.e.* normally unstable, non-inversion atmosphere) induces more intense pollution transport near the ground.

Discussion

In this paper we have reviewed the main sources of H_2S and SO_2 pollution and meteorological conditions at Kohtla-Järve, aiming to reveal the circumstances of their dependence. The atmospheric transport and diffusion of pollutants is influenced by a wide range of air circulations, thus it is complicated to match the exact emission source only on the basis of lowlevel meteorological data, which are strongly influenced by underling surface.

We have studied flux patterns with wind directions measured at 2–24 m heights, but also with those calculated for weather forecast purposes higher above. Wind directions at Kalevi monitoring station are quite different from those measured simultaneously at Aseri meteorological mast (Fig. 9). In addition to the systematic clockwise turn with height, there exist local "bends" within several ranges of wind directions. Wind shear due to surface



Fig. 9. Comparison of wind directions measured at Kalevi station at 3 m level and Aseri meteorological mast at 24 m level (2006–2009). Hourly data (gray) and moving average of 201 data points (or $1-3^{\circ}$ depending on direction) are given.

friction produces an ageostrophic component of the wind towards lower pressure. This is a result of balance between pressure gradient force, the Coriolis force, and the viscous force generally seen as turning of the wind in the boundary layer. According to the Ekman spiral, at the surface the wind is turned for 45° counter-clockwise compared to its direction in free troposphere [18]. In the real, vertically inhomogeneous atmosphere the direction changes usually less. It seems that the meteorological model HIRLAM, responsible for modeled boundary-layer wind directions rather than air quality model SILAM, is overestimating the wind shear angle, when downscaling from free troposphere to the surface layer. Thus, the wind at levels lower than 100 m is systematically turned counter-clockwise with respect to direct measurements.

Even two stations measuring at the same height and laying only three kilometres apart (Fig. 10) give strickingly different wind directions. The expected reasons are bending of wind trajectories around the semicoke

landfill and in much smaller scale, around the buildings (in location of Mobair station in particular). The effects of hills on wind field are known both in theory and measurement. Theoretical calculations with a model of atmospheric dynamics suggest significant wavelike disturbances a few kilometres downwind and also in lateral direction from 100–300 m high (horizontal dimension a few kilometres) hills [19].

Due to both the height-dependent and local effects, airborne pollutants should not necessarily originate from this exact direction, where the wind is blowing from. Detailed examination of multi-level wind measurement and also meteorological modeling data give us better imagination, where the pollutants come from. Topographically the dispersion of pollutants in the study area is determined by semicoke dumps, considerable industrial buildings and woodland belts, acting as microscale obstacles which form wakes.

The evaluation of correlations between different level wind directions was performed with available measured meteorological parameters. It was found that the airborne fluxes of sulphur dioxide from local sources in Kohtla-Järve (most probably VKG Oil) tend to be larger in intensely mixing atmosphere than under stagnant conditions. This may refer to the dispersion from elevated sources, from which emissions reach the ground easier in case of a well-mixed surface layer. The vertical wind standard deviations measured at the Aseri meteorological mast, which correlate with SO₂ concentrations, are a governing parameter of atmospheric dispersion [20]. Relations of H₂S fluxes with stratification parameters remain rather weak, thus no



Fig. 10. Comparison of wind directions measured at Kalevi and Mobair stations at 2 m level with those at Aseri mast, 24 m (29.11.2005–17.01.2006). Points represent hourly values, one-to-one (perfect fit) line is plotted for comparison.

clear conclusions on its releases can be drawn. Nevertheless, no evidence of highly elevated sources, in contrary to SO_2 , is found – in accordance with the hypothesis of wastewater treatment and semicoke dump sources.

Conclusions

The main achievement of the study is an empirical assessment of the effects of local- and microscale obstacles and atmospheric processes on diffusion of H_2S and SO_2 in the town of Kohtla-Järve.

- 1. The most enhanced sulphur dioxide and hydrogen sulphide levels at Kohtla-Järve (Kalevi) monitoring station originate from the direction of VKG Oil production units, semicoke dumps and thermal power plants of VKG Energia. Much higher concentrations measured at the territory of VKG Oil during MobAir episodic measurements suggest rather efficient dilution in the atmosphere when transported towards the town.
- 2. The secondary (but the most intense before 2008) peak of hydrogen sulphide originates from direction of the municipal wastewater treatment plant.
- 3. Sulphur dioxide measured at Kalevi station probably originates rather from elevated than ground-level sources, *e.g.* power plant stacks and industrial production units with height in tens of meters. Hydrogen sulphide, in contrary, may originate from lower-level sources.
- 4. Evidently the reconstruction of technological processes in both VKG units and the wastewater treatment plant have reduced the levels of sulphuric pollutants in the air of the town of Kohtla-Järve.

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