Time evolution of sulphur dioxide concentration generated by solid fuel combustion systems during the cold periods of the year

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Abstract.

About 99\% of sulphur dioxide present in the atmosphere comes from anthropogenic activities, the main source being industrial activity that processes sulphur materials, including burning fuels (coal, oil or gas), thus releasing sulphur dioxide into the air.

The purpose of this paper is to monitor the concentration of sulphur dioxide during two seasons, in the ambient air of Petroșani, with the aim of highlighting the possible atmospheric pollution caused by thermal power plants that use solid fuel during the winter period.

The monitoring periods were distributed over a period of 6 months (three in summer and three in winter), namely the summer months (May, June, July of 2022), the winter months of December (2022), January and February of 2023.

The main objective of the measurements is to identify if there is an increase in sulphur dioxide concentration in the atmosphere, resulting from the burning of solid fuels from household activities during the winter period.

Results indicate that there is a higher degree of SO\textsubscript{2} pollution in the winter period, but the recorded values are far below the limits set by national legislation. The paper presents some measures to limit sulphur dioxide pollution in the context of climate change.

Keywords: sulfur dioxide, atmospheric imissions, thermal power plants

1. Generalities

About 99\% of sulphur dioxide present in the atmosphere comes from human sources, the main reason being industrial activity that processes sulphur materials, including fuels (coal, oil or gas) and ores, thereby releasing sulphur dioxide into the air [1].

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The formation of sulphur dioxide from combustion of these fuels can be avoided if the sulphur compounds are removed before fuel is burned. Further oxidation of \( \text{SO}_2 \), usually in the presence of a catalyst such as \( \text{NO}_2 \), leads to the formation of sulfuric acid (H\( \text{SO}_4 \)).

Another mechanism is due to ultraviolet radiation from the upper atmosphere acting as a catalyst in the reaction between sulphur dioxide and oxygen, producing sulphur trioxide, which continues to react with water to form sulfuric acid or sulphurous acid.

\( \text{SO}_2 \) emissions from combustion taking place in natural gas boilers are low because the delivered natural gas is usually very low in sulphur.

Sulphur dioxide (\( \text{SO}_2 \)) resulting from the burning of solid fuels from stoves is a toxic and polluting gas that can have a harmful impact on the environment and human health.

At first, solid fuels such as wood and dry heat were used to heat homes and prepare food. However, soon, stove technology progressed and new stoves were introduced that burned coal, coke and tar. This caused more sulphur dioxide to be produced. However, many people still burn these solid fuels in their homes, causing local and even international pollution. [2]

Sulphur dioxide is found in air in small concentrations that do not pose a great risk to health, but these concentrations increase in the presence of solid burned fuels. This gas is harmful and can cause a large number of respiratory diseases, including asthma, an increased risk of allergies, headaches and breathing difficulties. At the same time, burning solid fuels can even contribute to global warming and atmospheric deconstruction. These intense combustions of solid fuels are among the main causes of high concentrations of sulphur dioxide in pollution. In order to reduce these high concentrations, we are experimenting with the introduction of more ecological alternatives for burning solid fuels and switching to even more renewable fuels. These changes can be made at governmental level and by educating the public on the ways in which they can contribute to reduction of polluting emissions.

Sulphur dioxide emitted by burning solid fuels can also have additional impacts on the environment, leading to water acidification and evaporation. Currently, emission factors of \( \text{SO}_2 \) are under control and surveillance in many countries, but certain emissions may persist for a longer period in certain regions of the planet. To reduce sulphur dioxide (\( \text{SO}_2 \)) emissions from burning solid fuels, governments can take appropriate measures, such as banning the burning of solid fuels, taxing the degradation of this fuel or providing subsidies for new technologies that can reduce pollution levels.

The largest producer of sulphur dioxide (\( \text{SO}_2 \)) emissions is the thermal energy industry. This sector uses solid fuels to produce electricity and thermal energy that is used in domestic and industrial applications [3]. A significant part of these solid fuels are coal and biomass, with wood chips and lignocellulosic materials being an important source.

Sulphur dioxide emissions can affect the population by worsening health problems such as chronic bronchitis and asthma. Exposure to sulphur dioxide can also cause inflammation of the airways and tissue inflammation. Another possible health problem is impaired kidney function. This is because sulphur dioxide forms ammonium and sulphates, which can negatively influence kidney function. Exposure to sulphur dioxide can also cause vision impairment and skin rashes.

Although there is a body of scientific research that points out how harmful sulphur dioxide emissions can be to humans and the environment, they continue to be used in energy production [4]. However, measures have been implemented to limit these emissions, such as the use of alternative fuels and the installation of emission control systems to reduce the amount of sulphur dioxide released, in order to keep pollution levels as low as possible.
2. Materials and methods

To measure sulphur dioxide emissions, the ECOTECH SERINUS 50 analyser was used, which has a photoluminescence (fluorescence) operating principle that involves the generation of an initial absorption phenomenon. It is made in accordance with the SR EN 14212:2012/AC:2014 standardized method of measuring sulphur dioxide concentration by ultraviolet fluorescence. The method is applicable to the determination of the mass concentration of sulphur dioxide present in ambient air up to 1000 µg/m³.

Luminescent molecules excited by certain quantum states (in this case by irradiation in the ultraviolet range), remain in this state for at least 10-9 sec, after which they return to the fundamental state, through luminescent emission. This secondary emission is proportional to the SO₂ molecules of sulphur dioxide in the sample and captured, transformed and transmitted as an electrical signal by the photomultiplier tube (PMT) of the apparatus. The fluorescence phenomenon occurs when the molecule, which is at a certain energy level, has at least two distinct electronic states whose level of rotation-vibration intersects in such a way that there is at least one level of rotation-vibration of the same value of energy, in each state.

The location chosen for monitoring was in the central area of Petrosani City, namely in the main building of INCD INSEMEX Petrosani. The sampling time for each set of measurements was 24 hours, being carried out three times in a month. These measurements were made in the yard of the main building of INSEMEX Petrosani, both due to the access to electricity supply and due to the positioning between the areas of interest, namely the Colonic District and the households on Maleia Street.

3. Results and discussion

In order to have a term of comparison if there is an increase in the concentration of sulphur dioxide in the winter period, measurements were made both in the summer and in the winter period. The measurements were made three days per month during 6 months including three months of the summer season and three months of the winter season. The main objective of these measurements is to identify if there is an increase in the concentration of sulphur dioxide resulting from the combustion of solid fuels from power plants using solid fuel during the winter period.

The measured values represent the hourly averages of the SO₂ concentration, and they were compared with the limit values set by law no. 104 of June 15th, 2011 regarding the quality of surrounding air.

Results of the measurements are presented in table no. 1.

Table 1. Monitoring of sulphur dioxide emissions in different periods of time.

<table>
<thead>
<tr>
<th>Date of measurement</th>
<th>Measurement hour</th>
<th>Hourly average SO₂ concentration, measured [µg/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>05.05.2022</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>09.05.2022</td>
<td>2.1</td>
<td>2.1</td>
</tr>
<tr>
<td>16.05.2022</td>
<td>2.1</td>
<td>1.9</td>
</tr>
<tr>
<td>07.06.2022</td>
<td>4.9</td>
<td>5.5</td>
</tr>
<tr>
<td>13.06.2022</td>
<td>6.6</td>
<td>6.6</td>
</tr>
</tbody>
</table>
According to table no. 1, all average hourly measurements fall within the limit values set by the national legislation. Also, the average concentrations of sulphur dioxide recorded during summer are significantly lower (1.8 times lower) than the average during winter.

Graphing the hourly concentrations for each month separately (fig. 1) shows that in the hourly periods 7-9 and 18-21 there are significant increases in SO2 concentration. These increases of the concentration of sulphur dioxide in certain hourly intervals (in winter) mostly coincide with the periods when fires are lit at thermal power plants on solid fuel. If solid fuel

<table>
<thead>
<tr>
<th>Date of measurement</th>
<th>Hourly average SO2 concentration, measured [µg/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>23.06.2022</td>
<td>2.0</td>
</tr>
<tr>
<td>04.07.2022</td>
<td>3.8</td>
</tr>
<tr>
<td>15.07.2022</td>
<td>1.1</td>
</tr>
<tr>
<td>25.07.2022</td>
<td>6.7</td>
</tr>
<tr>
<td>05.08.2022</td>
<td>4.0</td>
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<tr>
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</tr>
<tr>
<td>09.09.2022</td>
<td>2.8</td>
</tr>
<tr>
<td>20.09.2022</td>
<td>1.5</td>
</tr>
<tr>
<td>25.09.2022</td>
<td>1.1</td>
</tr>
<tr>
<td>06.10.2022</td>
<td>1.0</td>
</tr>
<tr>
<td>17.10.2022</td>
<td>1.3</td>
</tr>
<tr>
<td>23.10.2022</td>
<td>1.7</td>
</tr>
<tr>
<td>Average values for the summer months</td>
<td>1.7</td>
</tr>
<tr>
<td>Average values for the winter months</td>
<td>1.2</td>
</tr>
<tr>
<td>Daily average values during the summer period</td>
<td>8.144</td>
</tr>
<tr>
<td>Daily average values during the winter period</td>
<td>15.048</td>
</tr>
</tbody>
</table>

* Limit value for the mediation period of one hour according to Law no. 104 of June 15th, 2011 regarding the quality of surrounding air.
is burned uncontrolled, the amount of sulphur dioxide emitted is much higher than in regular combustion. The chemical reaction in this process can continue without the temperature, which can lead to much higher sulphur dioxide emissions than normal.

![Graphs showing hourly averages of SO2 concentration for different months.](image)

**Fig. 1** - Graphs of hourly averages determined on three different days of summer months on the left and winter on the right.

In order for the differences between the winter and summer periods to be more conclusive, the graph was made between the hourly averages in both seasons. According to figure no. 2, it can be observed that the winter concentration values are higher than the summer ones, but in both cases, they are below the limit of 350 µg/m³.
Fig. 2. Average concentration values in summer and winter.

The sulphur dioxide values recorded in winter are approximately twice as high as those recorded in summer. This fact leads to the conclusion that solid fuel combustion systems in nearby households have a significant influence on sulphur dioxide air pollution.

Air humidity increases in the morning due to condensation, when air temperature drops, air condenses around water particles in the atmosphere, which causes air humidity to increase. Soil can also absorb water from air at night, when heating from the morning sun causes it to evaporate into the air, increasing humidity. According to measurements made, an attempt was made to correlate the higher concentrations of sulphur dioxide in the morning with the increased atmospheric humidity, through the way it chemically reacts even in its gaseous state. This gas, along with others such as nitrogen dioxide, sulphur dioxides, and other sulphur compounds, can absorb moisture, which reduces the percentage of free gases in the atmosphere and, ultimately, their concentration. At low temperatures, gas condenses and turns into solid particles, which further reduces its concentration, so when atmospheric humidity is higher, there is likely to have a lower concentration of sulphur dioxide in the Earth's atmosphere. Under normal conditions, with the increase in humidity, the concentration of sulphur dioxide decreases in the morning, but in the present case the values indicate an increase. The only explanation for this fact is the increase in the concentration of SO$_2$ caused by spots in the nearby farming units. In order to heat houses, the inhabitants have to light the fire twice a day, once in the morning and once in the evening. These periods of increased SO$_2$ concentration coincide with the period of ignition of fires in individual solid fuel burning systems in households.

In the context of climate change, in order to limit sulphur dioxide pollution, a series of measures are suggested, which are presented below.

To reduce sulphur dioxide emissions, the best method is to use clean fuel and control combustion conditions, so as to avoid any contingents or excesses of fuel [5]. It is also recommended to replace solid fuel with alternative fuels, such as petroleum fuels or renewable energy.

Fuel selection, in the case of solid fuels, low sulphur fuels are preferred.

Using new efficient burners or moisture injection, and by using a well-adjusted and well-maintained plant [5].

The use of cleaning technologies, these include heat exchanges and electrostatic filters, air purifiers and other technologies that help reduce the concentration of sulphur dioxide. Actions to reduce uncontrolled burning of waste and fossil fuels.

Using these measures can significantly reduce sulphur dioxide emissions and contribute to a cleaner environment.
4. Conclusions

The purpose of the research is to monitor the concentration of sulphur dioxide in the ambient air in Petrosani, in two seasons, with the aim of highlighting the possible atmospheric pollution caused by the operation of thermal power plants and stoves that use solid fuel during winter.

Sulphur dioxide (SO₂) resulting from burning solid fuels from stoves is a toxic and polluting gas that can have a harmful impact on the environment and human health. Sulphur dioxide is found in the air in small concentrations that do not pose a great risk to health, but these concentrations increase when burning solid fuel. This gas is harmful and can cause a large number of respiratory diseases, including asthma, an increased risk of allergies, headaches and breathing difficulties.

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In order to have a term of comparison if there is an increase in the concentration of sulphur dioxide in winter, measurements were made both in the summer and in the winter period. The measurements were made three days per month during 6 months including three months of summer and three months winter. The main objective of these measurements is to identify if there is an increase in the concentration of sulphur dioxide resulting from the burning of solid fuels from stoves during winter.

Following the centralization of measurements, it resulted that all average hourly measurements fall within the limit values set by the national legislation. Also, the average concentrations of sulphur dioxide recorded during summer are significantly lower (1.8 times lower) than the average during winter.

Graphs of the hourly concentrations for each month separately, showed that in the hourly periods 7-9 and 18-21 there are significant increases in the concentration of SO₂. These increases of the concentration of sulphur dioxide in certain hourly intervals, in winter, are mostly due to the intervals in which fires are lit at the thermal power plants on solid fuel.

According to the measurements made, an attempt was made to correlate the higher concentrations of sulphur dioxide in the morning with the increased atmospheric humidity, through the way it chemically reacts even in its gaseous state.

At low temperatures, gas condenses and turns into solid particles, which further reduces its concentration, so when atmospheric humidity is higher, there is likely to have a lower concentration of sulphur dioxide in the Earth’s atmosphere. Under normal conditions, with the increase in humidity, the concentration of sulphur dioxide decreases in the morning, but in the present case the values indicate an increase. The only explanation for this fact is the increase in the concentration of SO₂ caused by spots in the nearby farming units. In order to heat houses, the inhabitants have to light the fire twice a day, once in the morning and once in the evening. These periods of increased SO₂ concentration coincide with the period of ignition of fires in individual solid fuel burning systems in households.

In the context of climate change, in order to limit sulphur dioxide pollution, a series of measures are suggested, namely:
- Use of clean fuel and control of combustion conditions, so as to avoid any contingents or excesses of fuel. It is also recommended to replace solid fuel with alternative fuels, such as petroleum fuels or renewable energy.
- Using new efficient burners or moisture injection, and using a well-adjusted and well-maintained boiler.
- Fuel selection, in the case of solid fuels, fuels with low sulphur content are preferred.
- Actions to reduce uncontrolled burning of waste and fossil fuels.

Acknowledgements

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References


