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Research concerning emissions monitoring by means of UV cameras

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Sulfur dioxide is one of the most abundant pollutants known in the world. It is emitted by antropic sources, especially due to the combustion of S containing fossil fuels, from both stationary and mobile sources. SO_2 emissions produce heavy pollution over areas with noticeable adverse health effects, especially in gaseous form or through reaction with condensed vapors, forming the acid rain. Volcanoes are responsible for a large amount of SO_2 natural degassing in the atmosphere, as well. Remote sensing measurement of pollution plume from distance is a new and challenging technique for air pollution monitoring. Thus stack emitted SO_2 plumes may be monitored without relying on the cooperation of the industrial facilities that generate the pollution. The SO_2 camera is a novel device developed using solar radiation scattered in the atmosphere as a light source for the measurements. The method is based on measuring the ultra-violet absorption of SO_2 in a narrow wavelength window around 310 nm, by applying a band-pass interference filter and a two dimensional UV - sensitive charge coupled device (CCD) detector. The paper is focused on real results accomplished in parallel at a Romania power plant, by using an UV camera for SO_2 emission, and the comparison with results obtained with a standard in stack measuring method. Based on the evaluation program developed using the measured values, the conclusion driven from the study is that the SO_2 concentration data delivered by remote sensing using UV cameras are appropriate to be used for stack emission evaluations. Still for getting an optimal correlation between emissions measured with standard methods, in stack, and remote sensing by UV cameras, one has to multiply the results, thus enabling to perform better corrections factors for the mathematical model used.

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1. Necessity of SO_2 monitoring

Presently, air quality is an important and global issue for all nations. The choice of a measuring process for air pollutants depends on the substance to be measured, the properties it has, and the information to be gained from the measured values. Measurements of main air pollutants must be carried out both at the location of its formation , thus determining the emissions' level and at different location, where the effect of the pollutant must be known, thus speaking about air quality, in respect to the particular pollutant' concentration in air. Sulfur dioxide (SO_2) is one of the most critical gases for the air pollution monitoring [2]. Sulfur oxides represent a major air pollutant and have significant impacts upon human health. SO_2 is considered also as a prominent greenhouse gas that contributes to global warming. The sulfur based oxides are responsible for a lot of respiratory and cardiovascular problems. Also sulfur oxides are known as precursor to acid rain and other atmospheric effects, with major toxicity and effects towards soil and vegetation [11]. The concentration of sulfur dioxide in the atmosphere influences the habitat suitability for plant communities as well as animal life, not at last upon human health. Anthropogenic SO_2 is almost exclusively formed during the combustion of fossil fuels with sulfur content and the most important representative energy source is fossil coal, because the sulphur content of other fuels, such as liquid and gaseous fossil fuels, can be easily removed before combustion [7] or is missing. Sulphur dioxide is an important gas for geologists and environmentalists, as it is known as having natural sources, as well, such as natural phenomena, of non-antropic origin. Volcanic SO_2 flux has been correlated with eruptions, but also explosions, forest burning, accidents, etc.

Different fuels are characterized by a wide range of sulfur content, most of which is released as sulfur dioxides, developed through combustion [5]:

- Oil and its by-products contain between 0.1 % by mass sulfur (paraffin) and 3-4 % by mass (heavy fuel oil) in the form of sulfides and thiols. Presently there are special commercial techniques to extract the sulfur and reduce its amount, thus the chance to generate SO_2 (ex. diesel) as well.
- Coal contains 0.1–4 % sulfur by mass, mainly as flakes of iron pyrites (FeS_2). The average sulfur content of European coal reservoirs is 1.7 % by mass, in Romania even more.
- Natural gas (known as being mainly methane CH_4) is normally sulfur free, but some gaseous fuels, according to their special origin or as byproducts of industrial processes, may contain also, up to 40 % by volume, hydrogen sulfide H_2S, that is generating SO_2 through combustion.

If the sulfur content (S) of the fuel is totally combusted, independent of its origin and bounds, SO_2 is formed through the general reaction [7]:

$$CH_3 - SH + 3O_2 \rightarrow SO_2 + CO_2 + 2H_2O \tag{1}$$

During the incomplete combustion, with lack of oxygen, elementary sulfur and hydrogen sulfide (H_2S) can be formed, at high temperatures, as well:

$$CH_3 - SH + 0.5O_2 \rightarrow H_2S + HCHO \tag{2}$$

$$2H_2S + O_2 \rightarrow 2H_2O + 2S \tag{3}$$

Hydrogen sulfide (H_2S) occurs during the combustion process of low quality coals like lignite or in the exhaust pipes of the piston engines exhaust gases, after their catalytic reduction, if the engine works at high load and low air-fuel ratio. During the combustion and in the exhaust channels, the SO_2 can be oxidized to SO_3 which will form, with the water from the flue gases sulfuric, the acid H_2SO_4. The SO_2 conversion to SO_3 is increased in the presence of vanadium of iron oxides at temperatures over 800 ºC, phenomena that is known as high temperature corrosion, specific to main combustion facilities (internal and external).

Evacuated into the atmosphere, SO_2 reacts in proportion of (1 – 2) ‰/ hour with oxygen, under the presence of ultraviolet radiation (*uvr*) and forms SO_3 [7]:

$$2SO_2 + O_2 + uvr \rightarrow 2SO_3 \tag{4}$$

Afterwards, the SO_3 will react with water vapors from the atmosphere to form sulfuric acid H_2SO_4. In periods of fog or days with high humidity the transformation rate of SO_3 to H_2SO_4 can be up to 15 % [7].

$$SO_3 + H_2O \rightarrow H_2SO_4 \tag{5}$$

Fig. 1 [7] presents an illustrated view of main atmospheric reactions for SO_2 and the global evolutions of the pollutant emissions into the air.

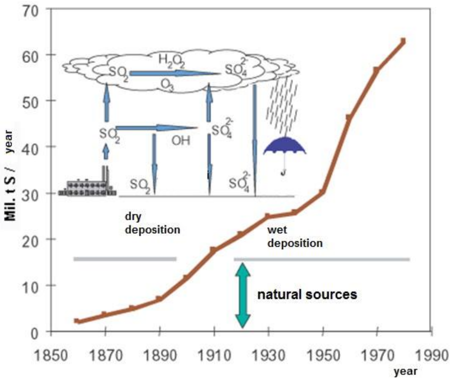

Fig. 1. SO_2 atmospheric reactions and emissions' sources [7].

For all mentioned reasons it is very important to monitor the concentration of SO_2 in the atmosphere, and especially control the sources (the larger ones), according to maximum admitted values in exhaust and further, generating trough dispersion, the air quality.

Remote sensing measurement for SO_2 was used successfully in the recent past for volcanoes activities and monitoring the exhaust emissions [3], [4]. Presently, this novel technique is proposed for power plants too. The technology is based on charge-coupled device (CCD) that is based on sensors capable to get information versus the UV portion of the spectrum [3] under the condition (hypothesis) that the camera is directed into the main central core of the stack flue gas plum, where the concentration of SO_2 is expected to be major.

This paper exposes the experimental results obtained from monitoring of the SO_2 concentrations, in comparison, exhausted by a large power plant located in Halanga, using simultaneous stack point measurement and remote sensing, with UV cameras. The comparative measurements are necessary as one of the main purposes of the research is to develop a method of a correct evaluating method for the level of emissions from stacks, by using only remote control. The method is also necessary for an overview or generating a prevention system of accidents or in case of exhausting of higher level of pollutants, which might be signalized and detected from distance, thus by remote control, by independent monitoring.

2. Experimental

2.1 Measurement location

Ambient air pollutants also have fluctuating concentration profiles, as pollutant emissions change constantly along with atmospheric dispersion conditions. Here the measuring method to be applied is essentially determined by the possible effect of the air pollutants. For long-term effects it is sufficient to determine mean values which can be fixed quite well by a series of individual discontinuous measurements [5]. To determine peak values, however, one must employ continuously operating measuring instruments, reflecting the temporal course of the concentrations as completely.

The Electric power plant Halanga (Fig. 2) is a main subsidiary company of the Romanian Nuclear Activities Authority (RAAN), located at 5 km North-East away from Drobeta Turnu Severin town, on 67 DN national route connecting Drobeta Turnu Severin and Tg. Jiu cities [6].

Fig. 2. Measurement location [11].

This means that the location is near the city Drobeta Turnu Severin, as well near agricultural sites (such as vineyards), or even the historical sites of the Romans, (for example the Traian bridge), sites that must be all preserved and protected from high pollution, especially generated by SO_2. According to its actual profile, ROMAG-TERMO Power Plant provides the process steam for ROMAG-PROD Heavy Water Plant (280 t/h and 31 MW) and also urban heating for Drobeta Turnu Severin Municipality (100 Gcal). In the same time, an electric power of over 160 MW is delivered to National Grid System [6]. The used Lignite (main fossil fuel used), is S rich, meaning that the SO_2 amount developed through combustion is significant. No desulphurization system is installed yet, even planned. All pollutants are thus leaving the furnaces through the stacks and reach the free atmosphere, as no other cleaning technologies for the flue gases are available, except particulate electro-filters [11].

2.2 Instrumentation

The literature [8], [9] describes a novel instrumentation called Vulcamera that is designed to operate with two cameras, simultaneously, with band pass filters centred on 310 nm and 330 nm. It is imperative to use two filters in these observations, to compensate for aerosol attenuation/backscattering, and this approach minimizes temporal mismatches associated with filter changes on a single camera [8]. [10] presents the tendency of doubling the results offered by cameras, near volcanoes sites. Thus the authors propose a comparison of UV camera-derived SO_2 emission rates with a mini-DOAS instrument.

Comparative measurements on which this paper are based occur, both for the stack emission control by means of a point measurement system (TESTO instrument working in amperometry), and the UV cameras (Fig. 3), that are generating remoting data about the concentration, from distance. Scope of using in parallel the two methods is:

1. Comparison of the values resulted and enabling thus the observing of the difference, if any, also the importance of the placement and peculiar conditions of the positioning of the US instrument;
2. Identifying of the program that is calculating according to the UV measurements the SO_2 concentration in stack, with satisfactory relevance.

Fig. 3. UV camera placed for the stack monitoring of the Halanga power plant [11].

The remote sensing instrument UV camera used during the research applies UV portion of the spectrum from 280-340 nm [10]. It has a high quantum efficiency detector and operates from a laptop. SO_2 camera is solely composed of a UV sensitive camera and a single spectral band-pass filter, allowing only radiation in a narrow wavelength interval encompassing significant SO_2 absorption structures to enter the camera optics [4].

2.3 Measurement technique for the UV camera

The preparing of measurement consists in some major steps which have to be followed. The first step is to connect camera to the power supply, next step is to connect it via USB interface to a laptop and finally give the command to cool down, assuring for example a level of cooled CCD by the limit of - 20ºC. Thus the electronic noise is reduced at its minimum. The user has multiple options for selecting the exposure time, adjust the numbers of measurements to take, what filters to use, etc. Cameras can be chosen to make measurements in automated mode using filter wheel. All the commands are given, under these circumstances, using PC interface [3].

Kantzas et al. [8] recommend a constructive solution with two Apogee cameras which offer more spatial resolution for observation.

The present proposed evaluation technique is based on a range of consecutive sequences, achieved both by standard methods (TESTO) and remote sensing (UV

Cameras). Firstly, the dark frame measurement is achieved and then calibration operation is undertaken, using two or more SO_2 cells with known concentrations on a portion with clear sky (nominated as A, B, in the particular case). Afterwards a clear sky measurement without SO_2 cells mounted was taken, in order to eliminate background noise.

In spectroscopy, the absorbance *A* is defined as [12]:

$$A_\lambda = \log_{10}(I_0 / I), \tag{6}$$

where: *I* is the intensity of *light(cd)* at a specified wavelength λ that has passed through a sample, meaning the transmitted light intensity,
I_0 - the intensity of the *light(cd)* before it enters the sample, or incident light intensity.

The apparent absorbance of SO_2 in the images of the plume is computed in correspondence to the apparent absorbance [1]:

$$\begin{aligned} &AA(i,j) = \\ &-\log\left[\frac{(PA_s(i,j) - PA_d(i,j)/(PA_b(i,j) - PA_d(i,j))}{(PB_s(i,j) - PB_d(i,j)/(PB_b(i,j) - PB_d(i,j))}\right] \\ &+ offset(i,j) \end{aligned} \tag{7}$$

where: *AA(I,j)* is the apparent absorbance at i^{-th} row and j^{-th} column pixel of the CCD,

PA and *PB* - intensity of image with filter A, respectively intensity of image with filter B.

The subscripts s, b, d represent in order: sample, background and dark images. Filter A corresponds to UV radiation absorbed by sampled plume, and filter B corresponds UV radiation unaffected by SO_2 molecules. An offset is removed if apparent absorbance of the clear sky is zero. Offset can be considered constant if the measurements are made in the midday when sunlight is relative constant. Conversion from apparent absorbance in ppm is made with SO_2 cells with known concentration.

Having as model a stand-alone, user-friendly code for measuring volcanic SO_2 fluxes, the utilisation of UV cameras was already applied. It consists of two elements: *Vulcamera_aq* and *Vulcamera_post*, which manage the image acquisition and all of the elements of post-processing, respectively in [9] it is presented that one has developed also an application for the calculation for the SO_2 concentration collected from UV cameras. In particular, the functions of the code include:

(i) Characterization of vignetting via the collection of clear sky images to generate a compensation for the angular dependency on pixel illumination; and

(ii) Determination of the calibration relationships between the absorbance value and the SO_2 cell concentrations, thus enabling conversion of the measured field images into ppm·m concentration maps [9].

In Fig. 4 the logic scheme developed is presented. It offers the possibility for the SO_2 concentration to be determined according to a novel simple program, developed accordingly with the above described principles. Firstly apparent absorbance is computed and one obtains a matrix *AA(I,j)*. After that, the concentration of SO_2 is computed by comparing intensities measured through two filters. Finally the program displays the SO_2 concentration according to the number of pixels.

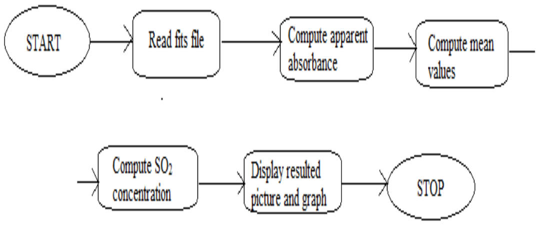

Fig. 4. Logic schematics for the calculation of the concentration of SO_2 estimated by means of US cameras' monitoring.

3. Results

According to the developed program of processing the data measured with the UV camera during a representative episode concerning the SO_2 emission from the stack of the power plant, following results were obtained.

In Fig. 5 the variation of SO_2 concentration using a filter A (310 nm) and a filter B (330 nm) accordingly with the number of pixels are represented. It can be observed that the maximum concentration appears in the middle of the plume. The central line (highlighted as red line) from the image represents the *j* column analyzed for SO_2 concentration. The number of pixels are represented mainly at right and on the bottom side of the image.

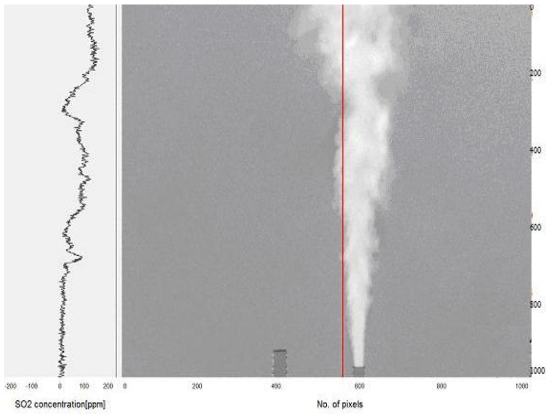

Fig. 5. Concentration of SO_2 estimated with the filters A and respectively B.

Fig. 6 represents the variation of SO_2 concentration, using another filter C (315 nm) and the same B filter (330 nm), accordingly with the number of pixels expected. It can be observed from Figs. 5 and 6 that for different plumes the concentration of SO_2 is different when the filter is changed. In the left side of Fig. 5 and Fig. 6 is represented variation of SO_2 concentration accordingly with the number of pixels. Maximum concentration of SO_2 is 163 ppm.

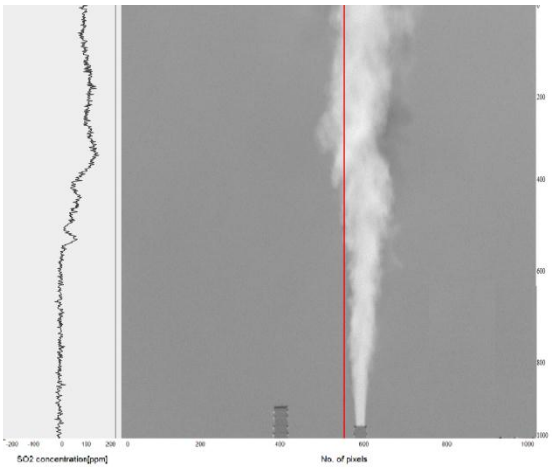

Fig. 6. Concentration of SO_2 estimated with filter B and C respectively.

For both Figs. 5 and 6 the concentrations are expressed in ppm.

4. Discussions

By comparing the case from Fig. 5 with that from Fig. 6, it results that the shape of the plume is different, and offers a very important information (depending of wind direction and speed), and that also the filters used for during the measurements, by equipping the cameras.

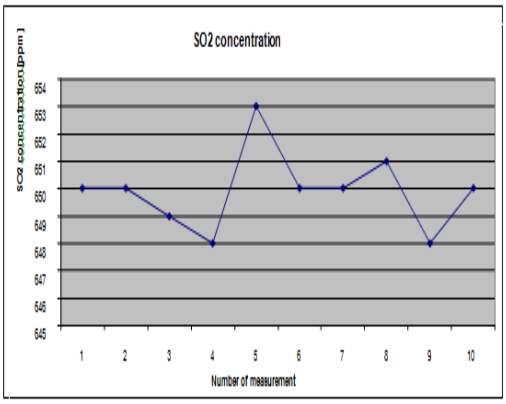

Fig. 7. SO_2 concentration measured in stack by classic standard method (TESTO).

Fig. 7 represents the variation of SO_2 concentration emission, measured with TESTO 350 XL, in the stack (using amperometry). The mean value is 649.9 ppm. The time interval between each sample of measurements is of 30 minute.

Comparative values between Testo classic method and UV camera data are presented, as resulted from the application of the computing program, by Fig. 8.

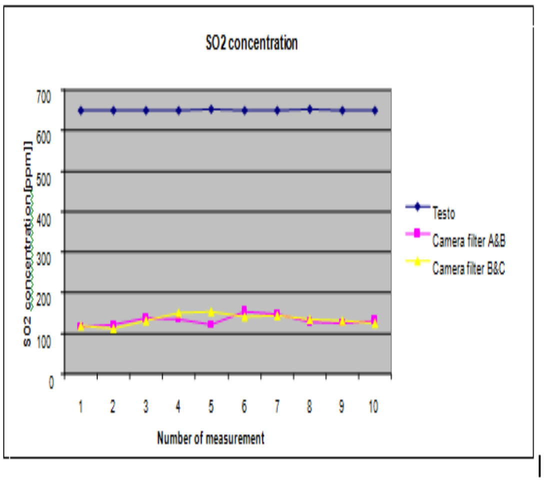

Fig. 8. SO_2 concentration generated in comparison by both techniques and different filters for the UV cameras.

Comparative measured data obtained with the Testo 350XL instrument and the UV camera, in comparison, is represented. It can be observed that values developed with Testo are four times higher than those generated by the camera (see Table 1). This four times discrepancy was found in other repeated series of measurements, achieved by the authors, on the same source, but under different episodes, as well, and based on this fact, the figure proposed is 4. But for sure, research must continue, until a consensus, under different conditions, is reached.

Table 1. Comparative values for SO_2 concentration [ppm].

	1	2	3	4	5	6	7	8	9	10
Testo	612	609	610	608	612	609	610	611	610	613
Camera filter A&B	142	128	135	131	163	160	124	146	159	157
Camera filter B&C	122	126	136	150	155	152	149	147	138	136

One notices, that by using different filter combinations for the cameras, no different results have been determined. The SO_2 concentration variation in this graph is thus concluding the existence of a quite important different for the case study selected, especially between the cameras' results and the stack monitoring technique. One considers that for getting camera data of comparative level with the stack measurements, a coefficient of multiplication (correction), in the used computational formula must be developed, so that the values generated by both techniques, in comparative simultaneous measurements should, at least, have the same, comparative level of ppm level of SO_2 concentration, meaning the same range with data measured with both Testo and UV camera. In order to verify the proposal, one appreciates that measurements and comparative analysis should be further accomplished. In the present case the coefficient is proposed to be around 4.

5. Conclusions

The paper focuses on results obtained with UV camera by remote sensing and the comparison with results obtained with stack continuous emission measurements. The conclusion is that both are offering data, but remote sensing is an option that is much more simple to be used, not being connected to the stack and offering thus a mobility and much more accessibility for external monitoring, as well.

Remote sensing technique is more and more becoming a future option for pollution measurement, not only for volcanoes, but also for man made emissions generated by power plants.

Up to now, UV cameras provided numerous benefits, such as high time resolution, which enables the capture of transient explosive events (such as volcanoes), the possibility to spatially resolve heterogeneous operations, e.g., fumaroles field sources and single-point operations. Furthermore, the camera images can be used to directly measure the plume transport velocity, potentially a major source of uncertainty in these measurements.

One concluded that, in this beginning stage, one has to introduce a correction factor for the data generated by the camera remote monitoring, in order to meet the expectations and range of the emission generated by standard methods, meaning in stack measured emissions. This necessity relies on several main causes, (i) the amount of SO_2, (ii) the dispersion of the plume, that does not always enable a correct centered remote control for the camera monitoring, meaning directly in the middle of the flue gas effluent, and (iii) also the variation by diffusion of the pollutant, in three directions, depending on the wind direction, turbulence and climate conditions. Both compared techniques are definitively based on different basics and phenomena; even they refer to the same pollutant, both have advantages and disadvantages. Remote controlling is applicable, with appropriate calibrated technique, as this novel method is presently developed by the authors, for external control as well, achieved not in stack, but from distance and might be used also as an alarming technique and/or included in a territorial accident prevention program.

Not at least artifact in the classic measuring techniques must be taken into account. Generally speaking, the proposed methods, even at beginning, must be further developed, but it is for sure that it might be used as alarming from distance method and a possibility to investigate from remote the sources of pollution, in this case the SO_2 flue gases emitted by antropic power plants that are running on sulfur containing fuels, with or filtering systems for the SO_2.

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