

Removal of Total Organic Carbon (TOC) from a Stationary Source using Diatomaceous Earth and Activated Carbon during the Processing of Polymeric Materials

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Abstract— Volatile organic compounds (VOCs) are among the most common pollutants released into the air in the petrochemical and chemical industries. The article deals with removing VOCs while producing plastic granules containing approx. 80% polypropylene and polyethylene, together with other additives. In this sector, the simplifying factor is that it deals with only one volatile pollutant, total organic carbon TOC, regardless of the specific materials from which the VOC emission originates. The permissible pollution level indicates a TOC emission limit of $50 \text{ mg} \cdot \text{m}^{-3}$ for this production. During operation, emissions are extracted from the machinery to a stationary source. It is a fabric filter where the air is cleaned through a layer of diatomaceous earth. To determine if the above limit is not exceeded, measurements showed that the TOC limits are significantly higher ($82.9 \text{ mg} \cdot \text{m}^{-3}$). Therefore, tests were carried out using a mixture of activated carbon and diatomaceous earth. The results showed that activated carbon significantly reduced TOC concentrations to $8.4 \text{ mg} \cdot \text{m}^{-3}$.

Keywords— Activated carbon, diatomaceous earth, polymers, stationary source, total organic carbon, TOC, volatile organic compounds, VOC.

I. INTRODUCTION

In the Czech Republic, according to the Air Protection Act of 2012, volatile organic compounds (VOCs) are any organic compound or mixture of organic compounds, except for methane. These compounds have the appropriate volatility under the specific conditions of their use (temperature $20 \text{ }^\circ\text{C}$, pressure 0.01 kPa and higher).

VOCs include most organic solvents, degreasers, lubricants, and fuels. The most common VOCs are methane, ethane, tetrachloroethane, methyl chloride, and various chlorinated hydrocarbons, the most common pollutants released into the air, especially by the petrochemical and chemical industries. Controlling VOC emissions is, therefore, significant from the point of view of environmental protection. VOCs in the air affect climate change, plant growth and decomposition, and human and animal health. Carcinogenicity studies have found that certain types of hydrocarbons can cause cancer [1].

VOCs are ubiquitous in indoor spaces, and several hundred organic volatile substances have been detected in indoor air by measurement. The characteristics of VOCs depend on their indoor or outdoor concentration in the air [2].

Carbon is dominant in the atmosphere in its inorganic form, carbon dioxide CO_2 [3]. Measuring total organic carbon (TOC) means measuring the amount of organic carbon in the air. Organic carbon compounds contain carbon with hydrogen, nitrogen, or sulphur bonds. They are not bound to carbonates. In some combustible systems, organic carbon compounds are contained in the fuel (wood, coal), which oxidize with the evolution of heat, carbon dioxide, water, and inert fractions (ash). In the presence of oxygen, the combustion of organics varies according to the nature of the material but can often begin to burn at $350 \text{ }^\circ\text{C}$.

A: Fabric filters and accessories

Fabric filters widely operate in the industry for cleaning discharged gaseous particles. They represent some of the most important devices used for environmental protection. They work to effectively remove industrial dust, especially its fine particles, or for the removal of VOCs, where adsorbents are also used. Fabric filters work as follows. The flowing air with dust particles passes through the filter, where the dust particles are trapped on the surface of the filter to form a so-called filter cake. The formation of a dust cake on commonly used filters is divided into three stages: deep, intermediate, and surface filtration. Cleaning the filter from the filter cake is usually done with compressed air inside the filter (regeneration) [4]. The principle of operation of the fabric filter is shown in Figure 1.

Fabric filters are usually part of a system to remove emissions from the production line. This system usually does not promise comprehensive removal of particulate matter or VOCs, but a well-designed system can prevent or reduce the likelihood of increasing emission limit concentrations.

In most cases, pipe ventilation is used in the system to remove VOC emissions or dust particles from the point of emission generation to a fabric filter (stationary source). The control system selection depends on the required quality of the discharged air according to the given legal regulations, especially the Air Act.

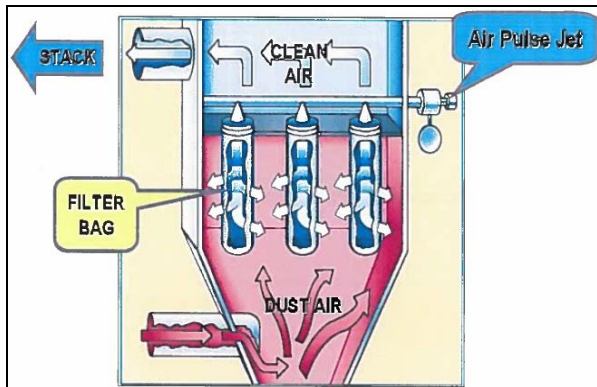


Figure 1: Fabric filter operation

A VOC or dust removal system can provide the required and effective control known as local exhaust systems. Safety requirements are given in the European Technical Standard EN 12779 [5]. A typical emission removal device consists of four components (Figure 2):

- Suction head for capturing particulate emissions from the source.
- Pipe system transporting emissions to the dust filter.
- Filter material that removes emissions from the air.
- Fan with sufficient power and energy [6].

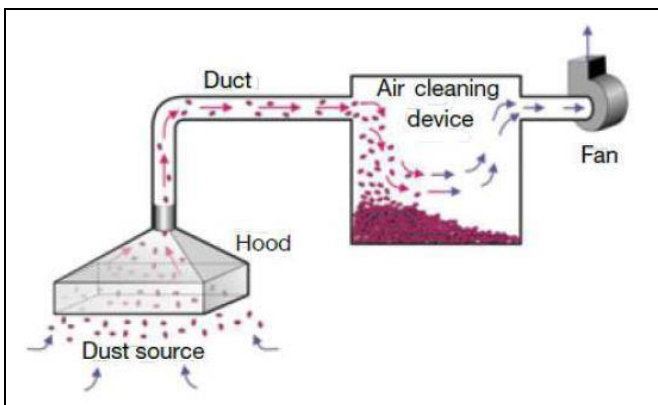


Figure 2: Dust removal system [6]

The filter is the essence of the fabric filter. Filtration performance affects the operational energy consumption and performance of the fabric filter [7]. In the past, filters were mostly made from a non-woven fabric whose goal was to create an effective porous barrier against pollutants. They were mostly materials of natural origin, e.g., felt. Currently, most filters are made from synthetic polyester and polypropylene materials in various grades and qualities [8]. Polytetrafluoroethylene (PTFE) is used as another synthetic material for filter production. Deqiang Jiang et al., 2008 compared the efficiency of a PTFE filter with a polypropylene needle felt filter, and the conclusions showed a 99.99% efficiency of the PTFE filter. Talc and limestone were used as test materials [9].

Screw extrusion machines or extruders are essential production equipment for processing semi-finished plastic products. They are essential for polymerisation and changing the properties of polymer substances necessary for producing the final product. The single-screw extruder was the first to be patented. I. G. Farbenindustrie patented a twin-screw extruder for reactive extrusion in 1940. A scientific article dealing with this method of polymer processing was first published in 1955 and was about the continuous esterification of cellulose. Reactive extrusion is widely used to modify polymers. Efforts to modify polyolefins with peroxides or a combination of peroxides with another monomer have been developing since 1960 [10].

Currently, twin-screw extruders are widely used in the plastics industry in a variety of applications. These are various processes that include mixing, blending, and reactive extrusion of polymeric materials. They are characterised by a perfect balance between the efficiency of mixing the mixture and the ability to create the required pressure. However, the values of the main input parameters must be known, including temperature, pressure, residence time, shear rate, and specific energy, at different positions along the screw profile. Furthermore, it is necessary to know how various factors affect, for example, the geometry of the screw, the phase of the speed or the speed of the screw [11]. Extruders consist of a hollow cylinder kept under stable internal temperature. They are equipped with an Archimedean screw-type screw with controllable speed. Therefore, the equipment of the extruder consists of a motor, a gearbox, heating elements, sensors, and a control system.

Polymer substances in a solid state (granules, flakes, dust) are usually delivered to the working space of the screw by gravity from hoppers. Another option is to supply the material to the feeding device at a prescribed speed. The material gradually moves along the screw and melts from heat conduction into a highly viscous melt. The melt is subsequently homogenised through dispersion processes and distribution mixing and is then forced through an extrusion head with a given number of holes and hole diameters (approx. 0.5 cm). From the extrusion head, the melt continues into the cooling bath, where it is again brought to a solid state. After this operation, the polymer material can be further processed.

The plasticization process in the inner part of the extruder is shown in Figure 3. The whole process consists of these individual operations known as functional zones:

- i) Supply of input material from hoppers.
- ii) Stage of pulling the material by turning the screw.
- iii) The phase of formation of a thin melt layer separates the material's solid component from the melt.
- iv) Gradual formation of melt.
- v) Moving the melt through the screw to the extrusion head.
- vi) Output of the melt from the extrusion head [12].

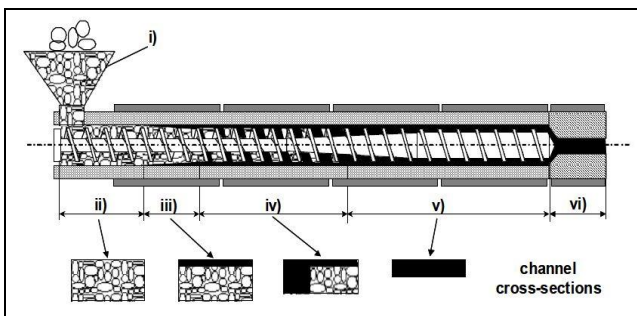


Figure 3: Melt production procedure in the inner part of the extruder [12]

Suction devices are used to capture harmful substances (gases, vapours, and solid particles) or excessive heat directly at the point of origin and thus prevent their spread into free space. Local suction is always more economical than general ventilation since suctioned airflow is always smaller than general ventilation.

If a pollutant with a mass flow of M_{ξ} [$kg \cdot s^{-1}$], leaks into the room, the airflow rate for the removal of the pollutant for general and local ventilation is determined by the relation:

$$V = \frac{M_{\xi}}{C_o - C_p}$$

where:

- C_o pollutant concentration in suction air [$kg \cdot m^{-3}$]
 C_p concentration of pollutants in the supply air (usually $C_p = 0$) [$kg \cdot m^{-3}$]

For total ventilation, the concentration of the pollutant in the suction air C_o must not exceed the permissible exposure limit C_{PEL} for the given pollutant, that is, $C_o \leq C_{PEL}$. For local suction, where the suction cover is appropriately designed so that the pollutant is not expected to enter the room, $C_o \gg C_{PEL}$ can be. The above relationship shows that the higher the concentration of pollutant in the suction air, the lower the suction air flow rate. The basic requirements for suction equipment are:

- Pollutants must be captured directly at the source with a purpose-designed suction attachment suitable for the operation and possible handling of the products.
- The air blown from the suction device into the outdoor environment must meet the legal emission requirements, and, if necessary, a separator (filter) must be included before the air is discharged into the atmosphere.

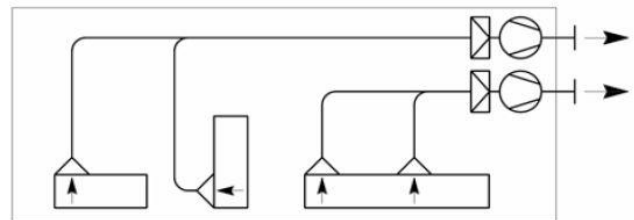


Figure 4: Diagram of group suction [6]

Suction systems are divided into the following:

- Central suction systems, where it is suctioned from several sources of the same or similar nature.
- Group suction systems (Figure 4) are used to suction one technological process, where substances that must not be mixed due to their danger are produced.
- Split suction systems use several fans connected to a technological process. They work mainly in large production lines.
- Unit suction systems are compact units containing a fan and a highly efficient filter to meet hygienic requirements.

A group suction system works for the selected technological process of polymer production. The system consists of two independent pipelines with separate fans. Each system is connected to a separate fabric filter. One system suction only dust from production, and the other warms waste air after the polymer melt enters the cooling bath.

Extraction systems consist of suction attachments, air ducts, a separator, a fan, and a suction pipe with a suction nozzle.



Figure 5: Suction attachments and suction slot for a screw extruder (extruder)

Suction attachments (Figure 5) capture pollutants at the source directly and should be designed as part of the technical equipment that produces the pollutants. They must be constructed so that, if possible, they surround the source of the pollutant. The cross-section through which the air flows into the suction attachment should be minimal, but it must allow air to flow at an acceptable speed, which varies from 0.5 to $2.5 \text{ m} \cdot \text{s}^{-1}$, depending on the harmfulness of the substance being sucked. If closed suction attachments cannot be used, e.g., for technological or handling reasons, a simple suction attachment must be placed as close to the source so that the flow of pollutants is directed into the attachment [13].

B: Description of the technology

The fabric filter under consideration works in a company processing polymeric substances. It involves mixing polypropylene, polyethylene, and other additives at a maximum of 65,000 tons per year.

The resulting product is polypropylene granules with modified properties for target customers. Customers also use pellets as input raw materials in equipment for producing plastic parts, especially for the automotive industry (bumpers, reflector chassis, ventilation distributions) or for further processing, e.g., plastic pipes, bottles, battery boxes, foils, etc. In addition, it is a processing activity of other synthetic polymers that permeates the entire production cycle. Figure 6 shows a diagram of the manufacturing process.

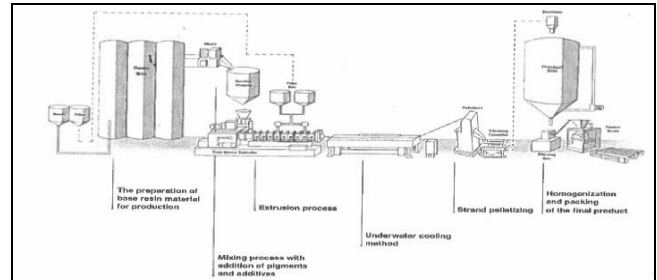


Figure 6: Drawing of production technology

Organic substances formed in small amounts during the extrusion of polymers can contaminate the product or the working environment. Therefore, they are sucked under intense pressure (0.8 to 1 bar) into a vacuum station. The vacuum station (Figure 7) contains an inlet vacuum drum with a diameter of 1000 mm and a length of 2150 mm, which is fitted inside with a labyrinth for separating condensed vapours of organic substances.



Figure 7: Vacuum station

It is followed by two parallel connected standing vessels with a diameter of 610 mm and a height of 1950 mm, which again have a dividing partition for better condensation of cooled organic vapours. The source of the vacuum is two vacuum pumps. According to the Waste Act, the separated condensate is handed over to an authorized person. The cleaned air is further led into a container with a diameter of 1100 mm and a height of 1230 mm with a dividing partition for the final condensation of cooled organic vapours and then fed into a stationary source, which is a fabric filter.

The air suctioned through the slots (Figure 5) at the end of the extruders is led to the filter unit, which is equipped with a fabric pocket filter (stationary source). Technically, these are two identical parallel connected independent filter units (Figure 8).

Diatomaceous earth is injected into the filter to increase the filter area and adsorb the residues of organic substances. Diatomaceous earth is dosed into the fabric filter manually through the dispenser. The manufacturer determines the dosage of diatomaceous earth at 200 kg per month. The fabric filter manufacturer guarantees 95% cleaning efficiency.



Figure 8: Filter unit with dispenser

Diatomaceous earth is a mixture of sediments consisting mainly of microscopic remains of shells of aquatic plants. The boxes contain mainly amorphous silicon dioxide SiO_2 . It is a very durable substance. Additionally, diatomaceous earth commonly contains carbonates and clay minerals, quartz, and feldspar [14]. The moisture content of diatomaceous earth is usually 50%, and the silica content varies between 86% and 94% [15]. Diatomaceous earth is very porous, light, chemically stable, and inert and is used primarily as a filter material or filler in the plastics industry [14].

Clean air is then released into the atmosphere through a 20-meter chimney. Separated diatomaceous earth is collected in the lower part of the filter unit, from where it is drained via a rotary valve into a container under the filter. The filter operation is alternated, so the filter layer can be renewed. The technical data of the fabric filter are shown in Table 1.

TABLE I:

TECHNICAL DATA OF THE FILTER UNIT

Nominal flow rate	$400 \text{ m}^3 \cdot \text{min}^{-1}$
Filter pocket size	$\varnothing 156 \times 3500 \text{ l}$
Number of filter pockets	162 ks
Filter surface	278 m^2
Compressed air	4 - 6 bar
Excipient	A mixture of diatomaceous earth

II. MATERIALS AND METHOD

The determination of the concentration of volatile organic substances is carried out by continuous measurement with a VAMET analyser before it enters the outdoor air according to EN 12619 [16]. The measurement also includes the determination of flow volumes of gas parameters and monitoring accompanying data on the operation of the measuring equipment according to technical standards EN ISO 16911- 1, EN 14790, and ISO 10780 [17] [18] [19].

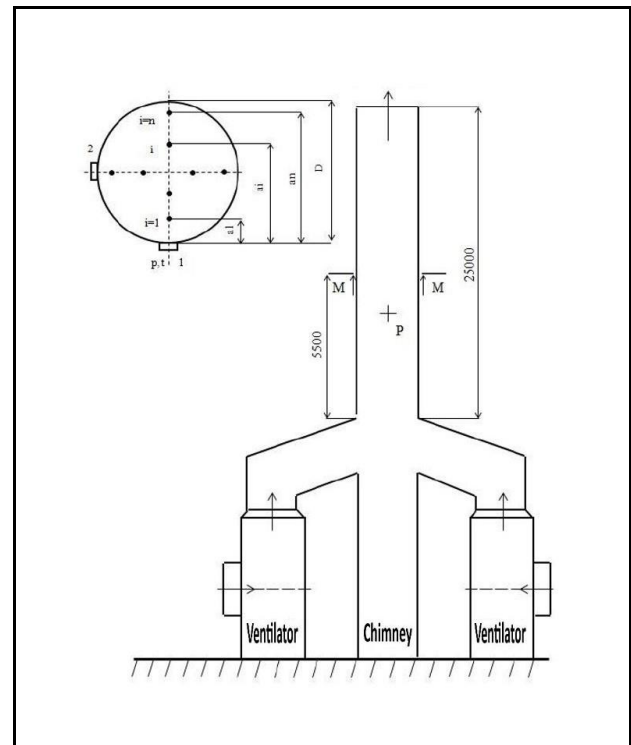


Figure 9: Scheme of emission measurement

The concentration of volatile organic substances under the normal state conditions of the warm gas is evaluated as mean values every 30 minutes of measurement. Therefore, twelve values of individual half-hour measurements are obtained. The average concentration of pollutants ($mg \cdot m^{-3}$) evaluates the ability of a stationary source (fabric filter) to comply with the emission limits, which is $50 mg \cdot m^{-3}$.

The emission mass flow of pollutants at the measuring point is evaluated from the average of individual measurements of flow volumes and individual measurements of concentrations of pollutants ($g \cdot h^{-1}$). Next, the specific production emission of the equipment is calculated as the ratio of the mass flow of emissions from the vents into the air to the amount of polymer substances processed. The specific production emission determines the emission of polluting substances from the equipment resulting from processing one ton of polymer substances. The emission measurement scheme is shown in Figure 9.

The measuring and sampling points for determining the concentration of pollutants behind the stationary source are located at the discharge of the suction fan in a chimney with a diameter of 1100 mm with a straight section of approx. 25 mm. A total of twelve measuring points for measuring the flow rate of hot gas are chosen in two perpendicular lines in the measuring section. The hot gas was taken from the point P, located on the axis of the pipe in front of the level of the measuring plane. The gauges used are as follows:

- AHLBORN humidity sensor
- ALMENO precision measuring device
- Humidity sensor
- Prandtl's tube
- TESTO digital pressure gauge
- Barometer
- Analyzer for continuous measurement of organic substances VAMET 145 with measurement range C_xH_y 0-1000 ppm
- COMET recording unit

Three repeated measurements measure flow volumes, static pressure, and humidity of hot gas according to technical standards EN 16911-1, ISO 10780.

The humidity of hot gas is measured according to the technical standard EN 14790 with a capacitive hygrometer and a precise measuring device ALMENO. The flow volume is measured with a Prandtl tube and a digital pressure gauge according to the technical standards EN 16911-1, ISO 10780.

The emission of the number of organic substances is determined according to the technical standard EN 12619 continuously using an analyser for the continuous measurement of organic substances VAMET using a teflon supply. The FID flame ionization detection method determines the concentration in a hydrogen flame.

Data on all continuous measurements performed are stored as instantaneous values in the memory of the COMET recording unit at minute intervals and stored for computer processing.

III. RESULT AND DISCUSSION

A total of three tests evaluated the amount of TOC emitted from a stationary source into the air. Test 1 (Figure 10) was performed under the standard conditions specified by the manufacturer. The stationary source contained 200 kg of diatomaceous earth, which was poured into the stationary source 24 hours before the beginning of the measurement, and the source was in operation.

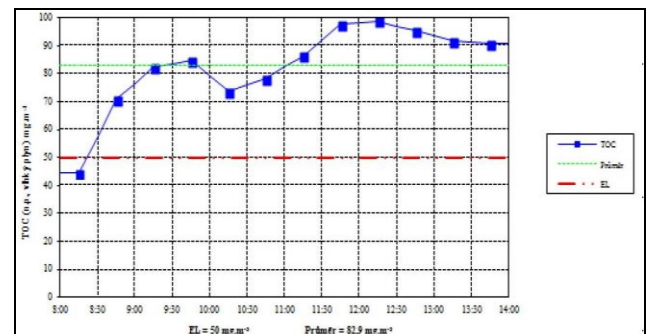


Figure 10: TOC measurement from a stationary source, test 1

Between 10:15 am and 12:15 pm, three individual measurements of static pressure, flow volumes, and humidity were made at the hot gas extraction outlet behind the stationary source. Between 8:00 am and 2:00 pm, a continuous amount of organic matter was measured at the exact location. The measurement includes twelve half hours of continuous measurement. The test results showed unsatisfactory results. The TOC concentration discharged from a stationary source was $82.9 mg \cdot m^{-3}$ (green line), which exceeds the established limit by 65.8%. In the period 11:45 to 12:30, it even approached the value of $100 mg \cdot m^{-3}$. Even the initial TOC concentration at 8:00 a.m. is relatively high, with a $45 mg \cdot m^{-3}$. Based on the above results, Test 2 was carried out when fresh diatomaceous earth (200 kg) was poured into the stationary source just before the start of the measurement. Figure 11 shows the result.

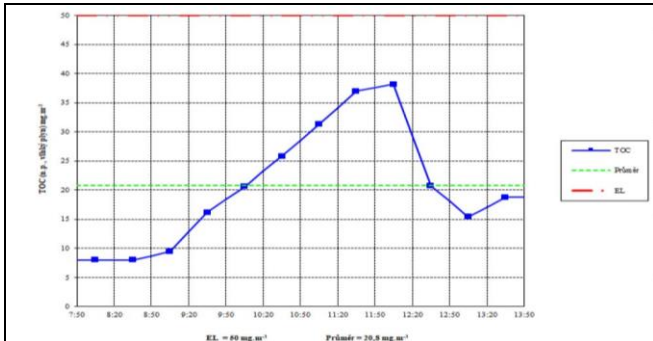


Figure 11: TOC measurement from a stationary source, test 2

Graph shows that the TOC concentrations at the beginning of the measurement using fresh diatomaceous earth were acceptable values. At 9:00 a.m., however, the TOC values began to rise, and 20 kg of activated carbon was poured into the stationary source at $38 \text{ mg} \cdot \text{m}^{-3}$. Values decreased substantially to an average concentration of $20.8 \text{ mg} \cdot \text{m}^{-3}$ (green line)

Activated carbon has a heterogeneous physical and chemical structure with micro, meso and macro pores in different dimensions, while its surface has non-polar and hydrophobic properties. It has a high adsorption capacity for capturing volatile organic compounds (VOC). It is not suitable for capturing polar substances such as ammonia NH_3 or sulfane H_2S . It is mainly made from coal, wood, shells, nuts, peat, or fruit kernels [20]. The results of test 2 show that with the addition of activated carbon, the TOC concentration values in the discharged air decreased rapidly. Therefore, the last test 3 was carried out, where the ratio of diatomaceous earth and activated carbon was mixed in a ratio of 2:1 (130 kg of diatomaceous earth) and 65 kg of activated carbon. The results are shown in Figure 12, where the average TOC concentration is $8.4 \text{ mg} \cdot \text{m}^{-3}$ (green line), which is significantly below the required limit for this type of stationary source.

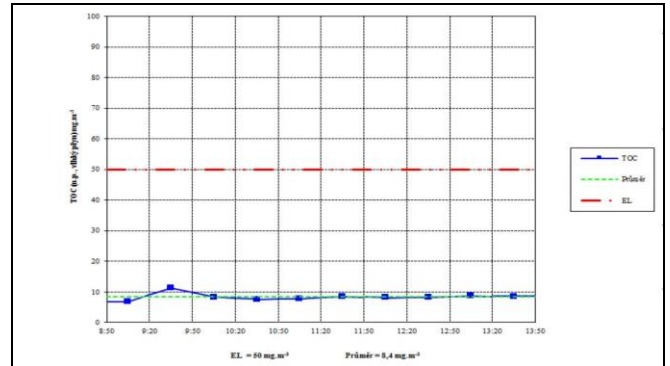


Figure 12: TOC measurement from a stationary source, test 3

IV. CONCLUSIONS

Spotless air has never existed in Earth's history. During the decomposition of plant and animal substances, forest fires, and animal death, the emissions of gases and solid substances became a standard part of life from the beginning. However, the technological development of humanity has caused a sharp increase in air pollution with consequences for people's health and the environment itself.

The Air Act requires all operators polluting the atmosphere of the Earth's air to comply with specified limits of total organic substances. For operators of stationary sources in technology processing polymers, the concentration limit of total organic carbon (TOC), which must not be exceeded, is $50 \text{ mg} \cdot \text{m}^{-3}$. The stationary source proposed in polymer technology processing a granulated mixture of polypropylene, polyethylene, and other additives proved unsatisfactory if only diatomaceous earth was used as an adsorbent. The first test showed that diatomaceous earth alone could not adsorb enough TOC to meet the given limits. The average TOC concentration during the first test was $82.9 \text{ mg} \cdot \text{m}^{-3}$, and in places, the concentration peaks approached $100 \text{ mg} \cdot \text{m}^{-3}$.

International Journal of Emerging Technology and Advanced Engineering

Website: www.ijetae.com (E-ISSN 2250-2459, Scopus Indexed, ISO 9001:2008 Certified Journal, Volume 12, Issue 10, October 2022)

After further tests, however, it became clear that activated carbon is capable of this activity, and the results showed a substantial reduction of TOC released into the air down to a concentration value of $8.4 \text{ mg} \cdot \text{m}^{-3}$. The ratio of diatomaceous earth and activated carbon to obtain this value was 2:1. TOC would undoubtedly be reduced even below this concentration if only activated carbon was used as an adsorbent. However, from an economic point of view, it is considerably more demanding. Activated carbon is roughly 2 to 3 times more expensive than diatomaceous earth.

V. FUNDING

This article was funded by the Ministry of the Interior of the Czech Republic from the Program IMPAKT1, Grant No VJ01010046 Innovation and development tools in the field of cause of fire investigation.

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