Research on Monitoring Method of Fuel Sulfur Content of Ships in Tianjin Port

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Abstract. Fuel sulfur content of marine sailing within the emission control area should not exceed 0.5%. Traditional boarding inspections are difficult, inefficient, and poorly targeted. Remote monitoring of air pollutants from ships is required to screen unqualified ships. The article investigates the topographical conditions and weather conditions of Tianjin Port, selects the technology of existing monitoring methods, elaborates and analyzes the composition and working principle of the current monitoring system, and finally forms a monitoring method of fuel sulfur content of ships in Tianjin Port.

1. Introduction
Since the proportion of ship trade and transportation has increased significantly, the atmospheric impact of ship emissions increased especially to port cities[1-2]. In 2016, the proportion of sulfur dioxide emissions from ships in Hong Kong was as high as 49% of the total local emissions. Based on this current situation, the International Maritime Organization IMO formulated the International Anti-pollution Convention MARPOL 73/78 Annex VI—"Rules for the Prevention of Air Pollution from Ships", requiring that the sulfur content should not exceed 0.5% after 2020[3-6].

The Ministry of Transport has adjusted the division of emission control areas and put forward new requirements. From January 1, 2019, sea-going ships entering the emission control area should use fuel with a sulfur content of not more than 0.5% m/m, large inland vessels and river-to-sea direct ships should use fuel that meets the requirements of the newly revised national standard for marine fuel oil. Starting from January 1, 2020, sea-going vessels entering the inland river control zone should use marine fuel
oil with a sulfur content of not more than 0.1% m/m. From March 1, 2020, ships that have not used alternative measures such as sulfur oxide and particulate pollution control devices entering the emission control area can only load and use marine fuel oil that should be used in accordance with the provisions of this plan. Starting from January 1, 2022, sea-going vessels entering Hainan waters in the coastal control zone should use marine fuel oil with a sulfur content of not more than 0.1% m/m.

The quality of marine fuel is uneven, leading to the discrepancy of the price. At present, there are still vessels entering the emission control zone using high-sulfur oil. Traditional methods such as checking oil change records and oil pumping monitoring are inefficient. Therefore, remote and rapid telemetry screening methods are urgently needed. At present, mature monitoring technologies, including sniffing technology, differential absorption spectroscopy technology and lidar technology have been widely used. Therefore, it is necessary to introduce marine fuel sulfur content monitoring technology to screen whether the fuel sulfur content exceeds the standard[7-10].

2. Ship Emission Monitoring Methods

Ship emission monitoring technology is basically divided into several categories: LIDAR, UV-CAM, Differential Optical Absorption Spectroscopy (DOAS), sniffer system, and portable multi-gas analyzer. Europe and the U.S started early, setting up emission control zones in ports and carrying out detection and screening of the fuel sulfur content[11]. In China, random inspections of marine fuel oil and document inspections are the main methods. Some scientific research institutes have conducted certain research on the technical methods that can be used to monitor and supervise the emission of air pollutants from ships.

The LIDAR technique is an active optical method where a short laser pulse is sent into the atmosphere. Part of the laser light is scattered back towards the instrument, this light is collected and analysed. The time delay between the emission of the light and its return to the instrument determines the distance to the source of the scattering[12]. A differential absorption LIDAR (DIAL) is capable of measuring the concentration of a gas in the atmosphere[13]. It does so by sending out pulses of two or more different wavelengths, chosen so that one wavelength is absorbed stronger by the gas to be measured than the other(s). The distance information along the path of the laser beam is still available, so the instrument determines the concentration at a known place in the atmosphere.

The UV-CAM exploits a strong absorption feature of the SO2 molecule in the UV region and is composed by a highly sensitive (between 280–320 nm) CCD array (1344×1024 pixels) manufactured and a UV transparent lens objective. The SO2 molecules being in the field-of-view of the camera cause attenuation of the recorded light intensity. By calibrating the camera using gas cells containing known amounts of SO2, the recorded light intensity can be related directly to the path concentration. Because the camera can sample rapidly (several images per second), features in the images can be tracked and the “in plume” wind speed and gas flux can be derived. The compact size of the instrument, the relatively low costs and the easiness of operation would make the instrument potentially attractive for routine monitoring of ship emissions of SO2[14].

The basic principle of DOAS is to use the differential absorption of light by gas molecules. In the vicinity of ultraviolet and near ultraviolet, the absorption spectrum of molecules is mainly caused by the electronic transitions of molecules or atoms. Various gas molecules have different differential absorption characteristics for light in different wavelength bands. For example, NH3 and NO have strong absorption near ultraviolet 200nm, SO2 and O3 have strong absorption in the 200nm ~ 350nm spectral range, and N2 is 440nm. The differential absorption nearby is very strong, and the absorption of CO is mainly concentrated in the infrared band. In this way, the absorption of different gas molecules can be determined and their concentration can be calculated.

Sniffer system is a method for simultaneous detection of pollutants under the atmospheric background concentration. When the plume of passing ships passes through the sniffer, the concentration of pollutants in the plume detected will be higher than the background concentration. The concentration of the pollutants discharged by ships can be obtained and the sulfur content in the fuel can be deduced.
There are many different types of portable multi-gas analyzer, the more commonly used is infrared gas detectors. The infrared detector has a wide range, high sensitivity and rapid response. After the gas to be measured is introduced, the light of its unique wavelength is absorbed, so that the incident infrared luminous flux is reduced. The higher the gas concentration, the less the luminous flux entering the infrared receiving gas chamber; and the luminous flux passing through the reference chamber and entering the infrared receiving gas chamber is constant. Therefore, the measured gas concentration can be calculated by the difference in luminous flux.

3. Method selection and analysis

The waterway of Tianjin Port is long and narrow, and ships enter the port successively. The port position is relatively concentrated, Environmental conditions such as ambient light, visibility, and wind speed are suitable. Through the analysis of existing technologies, it can be known that it is suitable to use sniffer technology, DOAS and LIDAR for monitoring experiments.

3.1 Sniffer system

Sniffer system is a method for simultaneous detection of pollutants under the atmospheric background concentration. When the plume of passing ships passes through the sniffer, the concentration of pollutants in the plume detected will be higher than the background concentration. The pollutant concentration can be analyzed and calculated, and the pollutant concentration discharged by the ship can be obtained and the sulfur content in the fuel oil can be inferred. The carbon content in the fuel used by ships is very stable, around 87%. Since the C and S elements in the fuel are fully combusted, most of the products are CO$_2$ and SO$_2$, and the remaining products are negligible due to the extremely low proportions. The sulfur content in the fuel is directly proportional to the SO$_2$ content in the plume, so the FSC and emission factors can be calculated from the ratio of the SO$_2$ to CO$_2$ concentration in the ship’s plume. The calculation formula is as follows:

$$\text{FSC} \% = \frac{S}{\text{Fuel} \text{ (kg)}} = \frac{SO_2 \text{ (ppm)}}{CO_2 \text{ (ppm)}} \times 32 \times 87\% = \frac{SO_2 \text{ (ppb)}}{CO_2 \text{ (ppb)}} \times 0.232$$

(1)

Since the concentration of SO$_2$ and CO$_2$ detected by the sniffing device includes the concentration of SO$_2$ and CO$_2$ in the atmosphere, the calculation is based on the concentration of pollutants in the plume, so the formula can be rewritten as:

$$\text{FSC} \% = \frac{(SO_{2\text{plume}} \text{ (ppm)}) - SO_{2BG} \text{ (ppm)}}{(CO_{2\text{plume}} \text{ (ppm)}) - CO_{2BG} \text{ (ppm)}} \times 87\% = \frac{\Delta SO_2 \text{ (ppb)}}{\Delta CO_2 \text{ (ppb)}} \times 0.232$$

(2)

Figure 1. CO$_2$ and SO$_2$ monitoring curves of the passing ships.
Sniffing technology may increase the concentration of pollutants in the ambient air during the measurement process. This may be caused by external factors such as surrounding power plants and factories. This type of concentration increase usually lasts for a long time, which is reflected in an increase in the overall background concentration. Only when the concentration of NOx and CO2 suddenly rises significantly within a period of time, can it be regarded as a ship plume passing by, and exhaust gas analysis and processing calculations should be performed. Actually, Not all fuel sulfur is converted to SO2. Depending on the combustion situation, 1% to 19% of the sulfur is converted to other forms such as SO3 and SO4. Therefore, assuming that all fuel sulfur is converted to SO2 will also have a certain impact on the actual sulfur content in the fuel, and part of the SO2 is converted to sulfate during the diffusion process, but the overall error does not affect the judgment of whether the ship uses low-sulfur fuel.

When using a sniffing system for actual monitoring, the influence of factors such as wind and ship position on the measurement results should also be considered. Therefore, it is necessary to comprehensively consider factors such as wind speed, wind direction and channel direction to determine the location of the monitoring point.

### 3.2 DOAS

DOAS has some advantages that cannot be achieved by traditional monitoring methods. The monitoring range of DOAS is very wide, and it can directly monitor a range of several square kilometers, so the measurement results are representative. The measurement period of DOAS is short, the response is fast, and a device can measure the mass concentration of several different gases at the same time, which is of great significance to the study of the changing discipline of pollutants emitted by ships.

During the measurement, the light source emits light with an intensity of $I_0$. After a certain distance of transmission, due to the different differential absorption of various atmospheric gas molecules, the intensity and structure of the spectrum will change accordingly. We set its intensity to be $I$, and the relationship between $I$ and $I_0$ can be derived from the Beer-Lambert:

$$I(\lambda) = I_0(\lambda) \exp\left\{ \sum_{i=1}^{n} \left[ -\sigma_i(\lambda) - \sigma_i'(\lambda) - \varepsilon_R(\lambda) - \varepsilon_M(\lambda) \right] N_i L \right\} + B(\lambda) \quad (3)$$

In this equation, $\lambda$ represents the wavelength, $\sigma_i(\lambda)$ is the measured narrow-band absorption cross section of the i-th gas molecule, $\sigma_i'(\lambda)$ is the broadband absorption cross-section, $N_i$ is the concentration of the i-th gas, $L$ is the optical path, and $n$ is the number of gas types to be measured, generally 2-10. $\varepsilon_R(\lambda)$ and $\varepsilon_M(\lambda)$ are the Rayleigh scattering coefficient and the $M$ scattering coefficient, respectively, and they change slowly with the wavelength. $B(\lambda)$ is the sum of various noises. Taking the logarithm of both ends of the equation (3), we can get

$$\ln \left[ \frac{I_0(\lambda)}{I(\lambda)} \right] = \sum_{i=1}^{n} \left[ \sigma_i(\lambda) + \sigma_i'(\lambda) + \varepsilon_R(\lambda) + \varepsilon_M(\lambda) \right] N_i L + B'(\lambda) \quad (4)$$

In actual measurement, formula (4) can be written as

$$R(\lambda) = \sum_{i=1}^{n} \sigma_i(\lambda) N_i L + P(\lambda) + B'(\lambda) \quad (5)$$

In the formula, $R(\lambda)$ is the natural logarithm of the ratio of the light source spectrum $I_0(\lambda)$ to the measured spectrum $I(\lambda)$, $P(\lambda)$ is a broadband spectral structure caused by Rayleigh scattering, $M$ scattering, detector response, broadband absorption of various gases, and the light source itself. As long as there are enough data points, the least square method is used for data processing, and the value of various gas concentrations $N_i$ can be obtained. Eliminate the incomplete coincidence between $R(\lambda)$ and molecular absorption cross-section $\sigma_i(\lambda)$ caused by small fluctuations in working environment temperature, working voltage and external environmental factors such as mechanical accuracy and vibration, these errors can be obtained by shifting, compressing and stretching the spectrum. The contrast value spectrum $R(\lambda)$ is fitted with multiple polynomials to obtain a fitting curve. The fitting curve can be considered as a slow change in the spectrum $R(\lambda)$. The fitting curve can be considered as a slow change in the spectrum $R(\lambda)$, which is, $P(\lambda)$.
from the spectrum $R(\lambda)$ can be regarded as the sum spectrum of the differential absorption of various molecules.

![Image](image_url)

**Figure 2. Using DOAS to monitor ship emissions. (SO$_2$SCD/molecules/cm$^2$)**

After noise reduction processing, the resulting ratio spectrum can be considered to be a spectrum containing only the sum of differential absorption of various molecules and noise. Since it contains the differential absorption of multiple molecules, the least squares method is used to invert their concentration at the same time, and the concentration inversion is realized through the method of data fitting.

The main sources of measurement errors in the DOAS are noise and the mutual influence of various gases. When the noise intensity is approximately twice the gas differential absorption intensity, the relative measurement error will exceed 10%, and the measurement result is not accurate enough. And during the measurement process of the DOAS, different gases in the atmosphere interfere with the measurement of a certain pollutant, which is related to their respective differential absorption cross sections. The more similar the differential absorption cross section to the pollutant, the greater the interference to the measurement. When multiple gases interfere with the measurement together, their interference effect is a linear superposition of the individual effects of each gas.

### 3.3 LIDAR

Lidar is based on the principle of Raman scattering generated by laser excitation of gas molecules in the atmosphere to detect the gas content in the atmosphere. In actual detection, in order to reduce the measurement error, the Raman scattering echo of nitrogen or oxygen molecules whose content in the atmosphere is stable and whose Raman backscattering differential cross section is accurately determined experimentally is used as the reference calibration value. The number of scattered photons is proportional to the number of molecules of the gas. By comparing the echo signals of the Raman backscattered light of the gas molecules to be measured and the reference gas molecules, the relative concentration of the gas to be measured can be obtained.

LIDAR emits two pulsed lasers with similar wavelengths to the same optical path in the atmosphere. One of the wavelengths is on the absorption line of the gas to be measured, denoted as $\lambda_{on}$, which is strongly absorbed by the gas to be measured; the other wavelength is on the edge or outside of the absorption line of the gas to be measured, and is denoted as $\lambda_{off}$, the gas to be measured absorbs little or no absorption to it. Since these two lasers have similar wavelengths, the extinction of other gas molecules and aerosols at these two wavelengths can generally be ignored. The difference in the echo intensity of the two laser beams is only caused by the absorption of the gas molecules to be measured, so the concentration of the gas molecules to be measured can be determined according to the difference of the echo intensities of the two wavelengths.

When a laser with wavelength $\lambda_i$ and pulse energy $E_i$ is emitted into the atmosphere, the number of echo photons $P_i(r)$ from the atmosphere with a distance $r$ and a thickness of $\Delta R$ received by the lidar can be obtained by the lidar equation:
\[ P_i(r) = \frac{E_i \beta_i(r) \Delta r A_i(r) \eta_1 \eta_2}{r^2 \left( \frac{c}{\lambda_1} \right)} \]
\[ \cdot \exp \left\{ -2 \int_0^R [\alpha_i(r) + N(r) \sigma_i] dr \right\} \] (6)

\( A \) is the area of the receiving telescope, \( \eta_1 \) is the quantum efficiency of the photomultiplier tube, \( \eta_2 \) is the efficiency of the receiving optical system, \( h \) is the Planck constant, \( c \) is the speed of light, \( N(r) \) is the concentration of contaminated gas molecules from the distance \( R \), \( y \) is the overlapping geometric factor of the emitted laser and the receiving field of view, \( \sigma_i \) is the absorption cross section of the measured pollutant gas molecule with a wavelength of \( \lambda_i \), and the atmospheric backscattering coefficient \( \beta_i(r) \) and extinction coefficient \( \alpha_i(r) \) include gas molecules \((M)\) and aerosol \((A)\)
\[ \beta_i(r) = \beta_{Mi}(r) + \beta_{Ai}(r) \]
\[ \alpha_i(r) = \alpha_{Mi}(r) + \alpha_{Ai}(r) \] (7)

If the selected two wavelengths \( \lambda_{on} \) and \( \lambda_{off} \) respectively correspond to the strong absorption and weak absorption of the measured gas molecule, the measured gas molecule concentration \( N(r) \) can be expressed as the following equation:
\[ N(r) = \frac{1}{2 \Delta \sigma} \frac{d}{dr} \left[ -\ln \frac{P_{on}(r)}{P_{off}(r)} \right] \]
\[ = \ln \frac{\beta_{on}(r)}{\beta_{off}(r)} - \frac{\alpha_{on}(r) - \alpha_{off}(r)}{\Delta \sigma} \] (8)

The difference in the absorption cross-section of the measured gas molecule is \( \Delta \sigma = \sigma_{on} - \sigma_{off} \); the atmospheric backscattering coefficients are \( \beta_{on} = \beta_{Mn} + \beta_{An} \) and \( \beta_{off} = \beta_{Moff} + \beta_{Aoff} \) respectively, the atmospheric extinction coefficients except for the measured gas molecules are \( \alpha_{on} = \alpha_{Mn} + \alpha_{An} \) and \( \alpha_{off} = \alpha_{Moff} + \alpha_{Aoff} \) respectively. Using the existing atmospheric model data, the pollutant differential absorption cross-section data and the \( P_{on}(r) \) and \( P_{off}(r) \) obtained by the differential absorption lidar, the distribution data of the measured pollutant concentration with the distance \( r \) can be obtained.

Figure 3. LIDAR measurements during the ship’s departure
The error sources of LIDAR measurement of pollutant concentration mainly include statistical errors caused by echo signal fluctuations, systematic errors caused by aerosol extinction and scattering, errors caused by the absorption of various gases, and errors caused by the selection of absorption cross-sections. Aerosols are widely distributed in the atmosphere and have complex optical properties, so it is easy to affect the measurement results. Since the absorption cross-sections of various gases overlap and influence each other, it is easy to cause interference. And the absorption cross section of different pollutant components is different in different environments, which also brings some influence to the measurement results.

4. Analysis of Tianjin Port Feature

Located at the western end of the Bohai Sea, Tianjin Port has two main waterways, Dagusha Channel and Dagang Port Channel, with a navigation capacity of 100,000 tons.

The wind direction of Tianjin Port is dominated by northwest wind in winter, and the wind is strong, and the maximum wind is above 8 levels. The southeast wind prevails in summer, and the wind direction is relatively unstable and the wind is weak. Strong convective weather brings thunderstorms and gales in summer from June to September. The normal wind direction is southwest wind, and the strong wind direction is northeast wind. The average annual wind speed is 4.2 m/s, which is relatively calm. It provides good conditions for the use of sniffing equipment.

There are few foggy days in Tianjin Port, and it is concentrated in autumn and winter. The average number of foggy days with visibility of less than 1km in the years is 16.5 fog days, and the actual number of days is 5.0, which generally does not hinder the navigation of ships. The number of available sunshine hours varies greatly in each season, with 1207.6 hours in spring and 15 hours in the longest day; 895.3 hours in winter, and only 9 hours in the shortest day. June and July can take the longest hours of photography, up to 450 hours per month; November, December, January, and February can take the shortest hours of photography, about 300 hours per month.

Tianjin Port's ships entering the port are mainly concentrated in Dagusha Channel and Dagang Port Area Channel. Tianjin Port has more ships entering the port on April, July and from September to December each year; less ships entering the port from January to March. The average daily number of ships arriving in the port in summer is 2 to 3 times that of winter. The daily arrival time is more average.

5. Monitoring Proposal of Tianjin Port

When drawing up a monitoring proposal, the characteristics, limitations, and measurement range of the equipment should be taken into consideration, combined with the characteristics of the port, and
analyzed to determine the location of the equipment. Since each technology has its disadvantages and the coverage of the monitoring range is not comprehensive enough, a combination of multiple technologies should be used for monitoring in the monitoring layout. Consideration should be given to light, climatic conditions, and regional openness, etc., and monitoring planning should be carried out in accordance with the actual situation of the port.

Taking into account the conditions of sunlight, wind direction, and mutual interference between different ships, a monitoring device is set up according to the actual topography of Tianjin Port. LIDAR has the largest detection range, so it should be installed between Dagusha Channel and Dagang Port Area Channel, so that it can simultaneously monitor the pollutant emissions of incoming ships. The detection angle of DOAS equipment can be adjusted as required, so it can monitor a large range of pollutant emissions from ships. The site selection principle is similar to that of LIDAR. The detection distance of the sniffer system is the shortest, and the measurement result is easily affected by the wind direction. Therefore, sniffer system should be installed at each end of the channel to ensure that the sniffing device can work normally under different winds.

Figure 5. Monitoring system installation location and monitoring range. Red represents LIDAR, blue represents DOAS, green represents sniffer system

6. Conclusions and outlook
There are a variety of monitoring methods for monitoring fuel sulfur content of ships. After investigating a series of environmental factors such as the topography, wind speed and illumination, visibility, and surrounding air conditions in the Tianjin Port area, sniffing technology, DOAS, and lidar technology are selected. When determining the suitable monitoring location, Tianjin Port’s geographical location, channel information, wind direction, temperature and humidity, visibility, seasonal light and other conditions should also be considered.

China will gradually extend the management area of the sulfur content of ships' fuel oil from the docks of typical ports to the entire emission control area. During this period, it is advisable to introduce as soon as possible the accumulation of technological research accumulated in the European and American emission control zones, and to serve my country's emission control zones as soon as possible.

Relying on typical ports to carry out experimental research on monitoring fuel sulfur content of ships, explore possible problems, such as the requirements for instrument accuracy and sensitivity, suitable detection distance and height, suitable wind and other meteorological conditions to provide standardized and extendable demonstration cases for other ports.

During the experiment, as a result of environmental factors and other conditions such as the number and frequency of ships entering the port in different seasons and periods, single or short-term monitoring results are accidental. Different equipment should be used to carry out long-term monitoring experiments on ships passing through each channel, and the location or status of the equipment should be adjusted according to the actual monitoring situation to ensure the accuracy of the results.
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