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**Remote sensing of sulphur dioxide emissions of
seagoing vessels**

D.P.J. Swart, A.J.C. Berkhout, G.R. van der Hoff,
J.B. Bergwerff and M.H. Broekman

Contact:

M.H. Broekman

Centre for Inspectorate Research, Emergency Response and Drinking Water

RIVM

e-mail: Marcel.Broekman@rivm.nl

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RIVM, P.O. Box 1, 3720 BA Bilthoven, telephone: 030 - 274 91 11; fax: 030 - 274 29 71

Abstract

Remote sensing of sulphur dioxide emissions of seagoing vessels

RIVM developed an instrument to measure sulphur dioxide emissions of seagoing vessels. In a five-day pilot study, the emissions of 24 ships on the Westerscheldt estuary were determined. As it turned out, a large number of those ships emitted huge quantities of sulphur dioxide.

Sulphur dioxide is a source of acidification and is harmful to the environment. Various measures have driven back emissions from other sources, such as traffic, industry and electricity generation. This causes the share of shipping in the total of the emissions to increase.

Seagoing ships are not allowed to use sulphur-rich fuel in territorial waters. This relatively cheap fuel may be on board, though, for use at sea. To what extent ship owners comply with this ban is not known. A breach is difficult to determine using traditional measurement methods because these require boarding the ship. The crew therefore knows a measurement is taking place and can adjust the type of fuel used.

The new technique is known as lidar (light detection and ranging) and measures from the shore. The lidar instrument uses a laser beam to scan the exhaust plume of a passing ship and to determine the emission, unnoticed. An advantage of this method is that nearly every passing ship may be measured, instead of only a few.

On land, sulphur dioxide emissions of industrial installations are limited by licences. These are granted on the lines of the Dutch emission guideline air (NeR, April 2003), which puts demands on sources that emit more than 2 kg per hour (0.56 gram per second). The emissions of all measured ships turned out to be higher than that. The highest emission measured was 36 gram per second. This indicates the importance of recognising ocean shipping as a source of air pollution, both when issuing rules and when enforcing them.

Key words: sulphur dioxide, SO₂, emission, ocean shipping, lidar, remote sensing

Rapport in het kort

Zwaveldioxide-uitstoot van zeeschepen op afstand gemeten met lidar

Het RIVM heeft een instrument ontwikkeld om de zwaveldioxide-uitstoot van zeeschepen te meten. In een proefstudie van vijf meetdagen werd voor 24 schepen op de Westerschelde de uitstoot bepaald. Een groot aantal daarvan bleek forse hoeveelheden zwaveldioxide uit te stoten.

Zwaveldioxide is een bron van verzuring en is schadelijk voor het milieu. Diverse beleidsmaatregelen hebben de uitstoot van andere bronnen van zwaveldioxide, zoals verkeer, industrie en elektriciteitsopwekking, flink teruggedrongen. Het aandeel van de scheepvaart in de totale uitstoot wordt daardoor steeds groter.

Zeeschepen mogen binnen de territoriale wateren niet op zwavelrijke brandstof varen. Deze relatief goedkope brandstof mag echter wel aan boord zijn voor gebruik op zee. Het is onbekend in hoeverre reders zich aan dit verbod houden. Met traditionele meetmethoden is een overtreding moeilijk vast te stellen aangezien deze metingen aan boord plaatsvinden. De bemanning is daardoor op de hoogte van de meting en kan het stookgedrag aanpassen.

De nieuwe techniek heet Lidar (*light detection and ranging*) en meet vanaf de wal. Het lidarinstrument scant met een laserbundel de rookpluim van een passerend schip en stelt zo onopgemerkt de uitstoot vast. Een voordeel van deze methode is dat nagenoeg elk voorbijvarend schip kan worden gemeten, in plaats van slechts enkele schepen per dag.

Op het vasteland worden zwaveldioxide-emissies van industriële installaties beperkt door vergunningen. Deze worden verleend aan de hand van de Nederlandse emissierichtlijn lucht (NeR, april 2003), die nadere eisen stelt aan bronnen boven de twee kg zwaveldioxide per uur (0,56 gram per seconde). De uitstoot van de gemeten zeeschepen bleek daar in alle gevallen boven te liggen. De hoogst gemeten uitstoot bedroeg 36 gram per seconde. Aandacht voor de zeescheepvaart als bron van luchtverontreiniging is dus van belang, zowel bij regelgeving als bij handhaving.

Trefwoorden: zwaveldioxide, SO₂, emissie, zeescheepvaart, lidar, remote sensing

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Summary

This report describes the use of a new measurement method for determining the sulphur dioxide emissions of seagoing vessels. The measurements were made from an inspection vehicle on shore using a scanning laser beam. This technology is called LIDAR.

This method can be used by the VROM Inspectorate when monitoring the sulphur content of fuels, both as an independent instrument and in combination with other methods. The most important advantage is that the ship's crew is unaware that measurements are being conducted. Another advantage with respect to more traditional methods is its efficiency: virtually every passing ship can be measured. When the lidar technology is combined with other methods, the lidar measurements can be used to determine which ships should be boarded for additional testing with the other methods. In this case, the lidar is used as a surveillance and detection instrument.

In 2006, a pilot study was conducted on the Westerscheldt estuary. On 5 measurement days, the emissions of 24 ships were determined. For all 24 ships, the observed emission was higher than 0.56 g of sulphur dioxide per second, the limit value above which the Netherlands Emission Guidelines for Air (NeR, April 2003) places additional demands on the emissions of industrial installations on land. The highest measured emission was 36 g per second.

As part of the study, the lidar instrument was adapted so it could be used to detect sulphur dioxide and measure the moving smoke plumes of the ships. These developments were successfully completed and resulted in an operational instrument. Based on the pilot study, a lower limit of quantification of 0.1 g per second was established. The emissions of all ships measured were far above this level. A typical emission measurement has a measurement uncertainty of approximately 20%.

During the study, it became clear that the wind direction plays an important role in the usability of the lidar method. From a single measurement location, measurements can be made only for a limited number of wind directions. In the future, it will therefore be important to be able to work from multiple locations that are suitable for various wind directions.

1 Introduction

Seagoing vessels are an important source of sulphur dioxide in the atmosphere. There are two reasons for this. Firstly, the contribution of other sources, such as electricity generation, industry and traffic, is declining due to stricter legislation. Secondly, the sulphur fraction in heavy marine fuels is rising. These causes are linked together. Due to the stricter "onshore" requirements, a steadily increasing proportion of the sulphur in crude oil is finding its way into marine fuels.

1.1 Norms for sulphur dioxide emissions

On land, the sulphur dioxide emissions of industrial installations are limited by means of permits. These permits are based on the Netherlands Emission Guidelines for Air (NeR, April 2003). These guidelines place additional demands on sources with emissions above 2 kg of sulphur dioxide per hour (0.56 g per second). These sources are required to limit their emissions to no more than 50 mg per cubic meter of flue gas by means of purification measures. Nevertheless, within this norm significant emissions are possible on land. The ten largest sulphur dioxide sources on land have emissions that are above 100 g per second¹ (approximately).

There are currently no norms in the Netherlands or other countries that apply to the atmospheric emissions of hazardous substances in the flue gases of seagoing vessels (Broekman, 2006). However, demands are placed on the fuels being used.

1.2 Norms for the sulphur fraction of fuels

The maximum allowable quantity of sulphur in marine fuel depends on the type of fuel and the location where the fuel is used. Moreover, several norms will become stricter in the near future.

On European waters, a maximum sulphur content in gas oil² of 0.2 percent by mass applies until the end of 2007 in accordance with EU Directives 1999/32 and 2005/33. From 1 January 2008, a maximum sulphur content of 0.1 percent by mass will apply to gas oil.

Until the end of August 2007, diesel and fuel oil used on the North Sea/Channel have been subject to a norm of 4.5 percent sulphur by mass. From the end of August 2007, diesel and fuel oil used in those areas will be subject to a norm of 1.5 percent sulphur by mass. At a previous stage, this change was applied to the Baltic Sea (at the end of August 2006). This means that from the end of August 2007, the stricter norm for diesel and fuel oil (maximum

¹ derived from data in the Dutch Emission Inventory for 2004.

² The definitions of the various types of marine fuels are listed in Article 2 of EU Directives 1999/32 and 2005/33.

sulphur content of 1.5 percent by mass) will apply to a contiguous area including the North Sea/Channel and the Baltic Sea.

On the open sea outside the defined areas (North Sea/Channel, Baltic Sea) a maximum sulphur content of 4.5 percent by mass applies to all marine fuels.

1.3 Problem definition

Sulphur-rich fuels are significantly cheaper than sulphur-poor fuels. Ships can carry sulphur-rich fuels on board, but they cannot be used within territorial waters. However, enforcing this rule is difficult if monitoring can only be done on board. It is therefore conceivable that sulphur-rich fuels are being used on major shipping routes such as the Westerscheldt. In that case, the emissions are being significantly underestimated. A suitable enforcement instrument is lacking.

1.4 Aim of the project

The aim of the project is to investigate whether, and to what extent, the above problem can be solved by using lidar technology, and whether an accurate picture can be obtained of the sulphur dioxide emissions of seagoing vessels on the major shipping routes in the Netherlands.

This technology has the important advantage that the measurements can be conducted remotely, and therefore go unnoticed. The instrument works with a laser beam and is a type of radar for sulphur dioxide with a range of approximately 2.5 km. With this lidar method, the emissions of seagoing ships can be measured while they are underway.

The RIVM has developed and built this lidar system in cooperation with a number of external parties, including the VROM Inspection and Investigation Service and the National Police Services Agency. It is a mobile instrument that specifically focuses on measuring emissions remotely to benefit surveillance and enforcement. The instrument is carried in an inspection vehicle that provides all necessary infrastructure and can operate entirely independently. At the present time, this mobile lidar is capable of measuring concentrations and emissions of three trace gases: SO₂, NO₂ and NH₃.

In 2005, the instrument was first used operationally to evaluate satellite measurements of NO₂. It was used again for this purpose in 2006. In 2006 it was used on behalf of the VROM Directorate for Climate Change and Industry to conduct the first operational emission measurements of NH₃; this was initially done on an artificial source, and then on an actual source (a fertilized pasture). These measurements were all successful and will be continued in 2007. The list of detectable gases will possibly be expanded with NO and/or benzene.

Figure 1-1 shows the exterior and interior of the inspection vehicle.



Figure 1-1. The inspection vehicle, exterior en interior.

1.5 Research question and realization

There were two parts to the research presented in this report, both of which were realized:

- (1) Making the lidar suitable for measuring the SO₂ emissions of seagoing ships while underway .

Important technical challenges in this part of the research were scanning the smoke plume with the laser beam and analyzing the measurements with a very short integration time. Both modifications were necessary because the ships were in motion, so there was not much time to conduct the measurements. The technology used is described in Chapter 2.

- (2) Conducting a pilot study, where the emissions of ships were measured while they were underway.

In the pilot study, measurements were conducted on the Westerscheldt during five days in total. We attempted to measure the emissions of 42 passing ships. These attempts were successful in 24 cases. The results are presented in Chapter 3.

Chapter 4 discusses the results and addresses a number of characteristics of the measurement technology that are important with respect to enforcement, such as precision and selectivity. A number of conclusions and recommendations are presented.

2 Materials and Methods

2.1 Lidar technology

The acronym lidar stands for *light detection and ranging*. This technology has many similarities with radar. A brief pulse of light is emitted. Some of the light is reflected by molecules and aerosols in the air. This reflected light is received with a telescope, detected and analyzed. By measuring the time lapse between sending and receiving the light, the distance to the reflecting particles can be derived.

The lidar system used in the present study sends out two differently coloured pulses of light in rapid sequence. The colours are chosen in such a way that the first colour is more strongly absorbed by the target gas (in this case SO₂) than the second colour. If SO₂ is present, the reflected light from the first light pulse will be more strongly attenuated than the light from the second pulse. The SO₂ concentration at the location where the light is reflected can be derived from the degree of attenuation. Because molecules that reflect light are present everywhere along the route of the light beam, it is theoretically possible to also determine the concentration along the entire route. In practice, however, a value can be determined every 100 to 200 m, from about 350 m to about 2500 m from the instrument.

By making such a concentration measurement in the same horizontal direction, but by varying the vertical direction, the concentration distribution of SO₂ can be determined in a vertical plane. This is shown schematically in Figure 2-1.

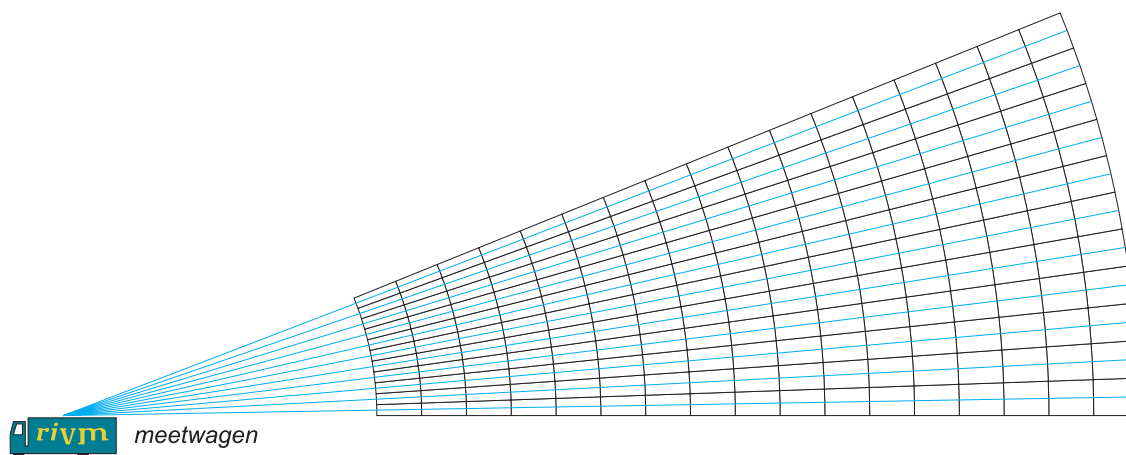


Figure 2-1. Schematic overview of the determination of the SO₂ concentration in a vertical plane. The measurement directions are shown in blue, the black cells indicate segments for which a concentration is determined. **Meetwagen = inspection vehicle**

For the emission measurements of the seagoing vessels, a vertical plane was used that was composed of 9 directions. The maximum distance was approximately 2.5 km, the maximum

elevation about 300 m. Measuring all directions in a scanning plane takes about 45 seconds, after which the light beam is returned to the initial position and the scanning plane is again measured. In principle, such a cycle can be repeated an unlimited number of times.

2.2 Determining the emission

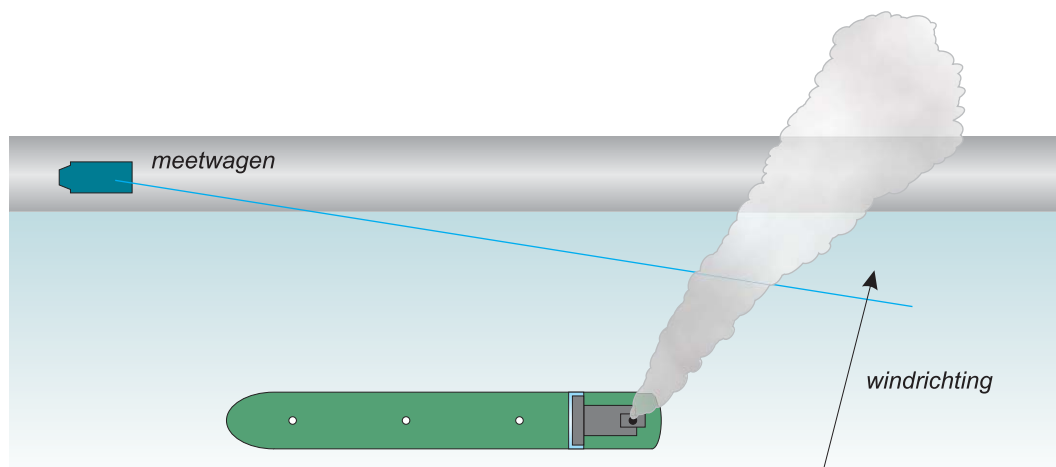


Figure 2-2. View from above of the situation during an emission measurement.

Meetwagen = inspection vehicle **Windrichting = wind direction**

Figure 2-2 is a schematic representation of how the emission is measured. The lidar is set up on shore. The vertical scanning surface is located as much as possible at right angles to the wind direction and parallel to the direction the ships are travelling. The instrument is turned on and begins to measure SO_2 concentrations continuously. If a ship passes, the smoke plume is driven by the wind through the scanning plane (Figure 2-3).

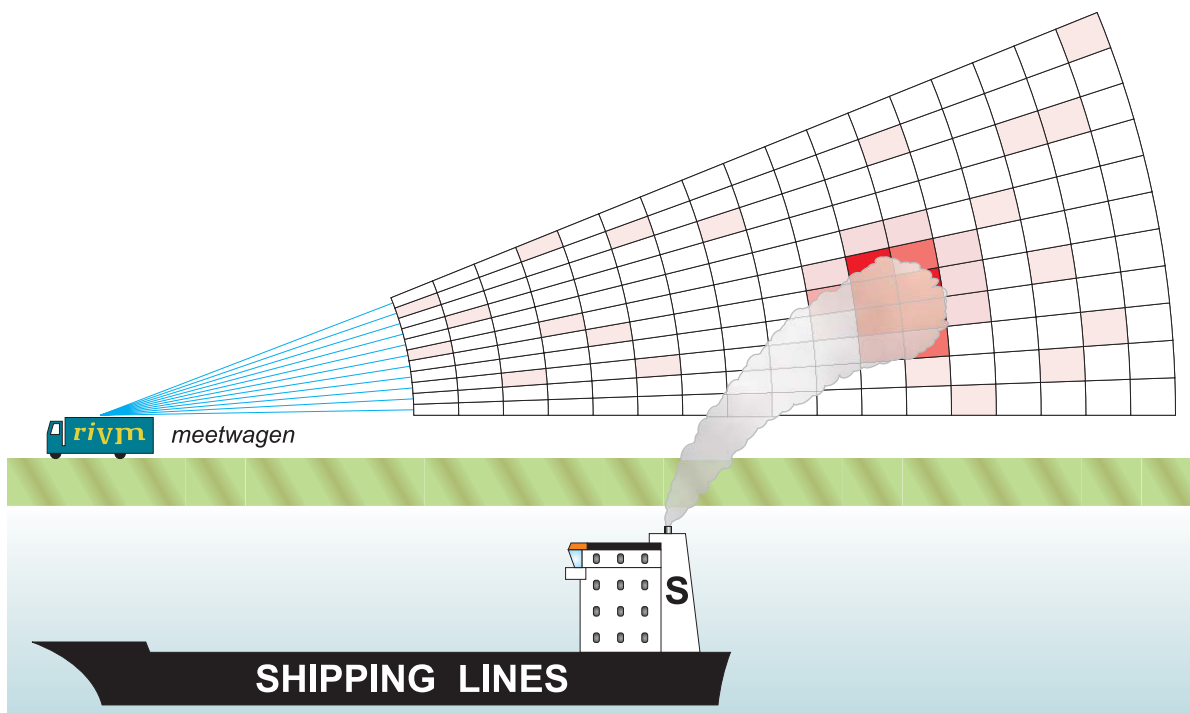


Figure 2-3. View from the side of the situation during an emission measurement.

Meetwagen= inspection vehicle

In the lidar signal, the soot and other particular matter in the smoke plume can be seen. In this way, it can be determined where the plume passes through the scanning plane. At the same location, the SO₂ concentration is determined. The area of the section through the plume can also be derived from this information. Finally, to determine the emission factor, these two results – the concentration and the area – are multiplied by the wind speed.

2.3 Measurement procedure

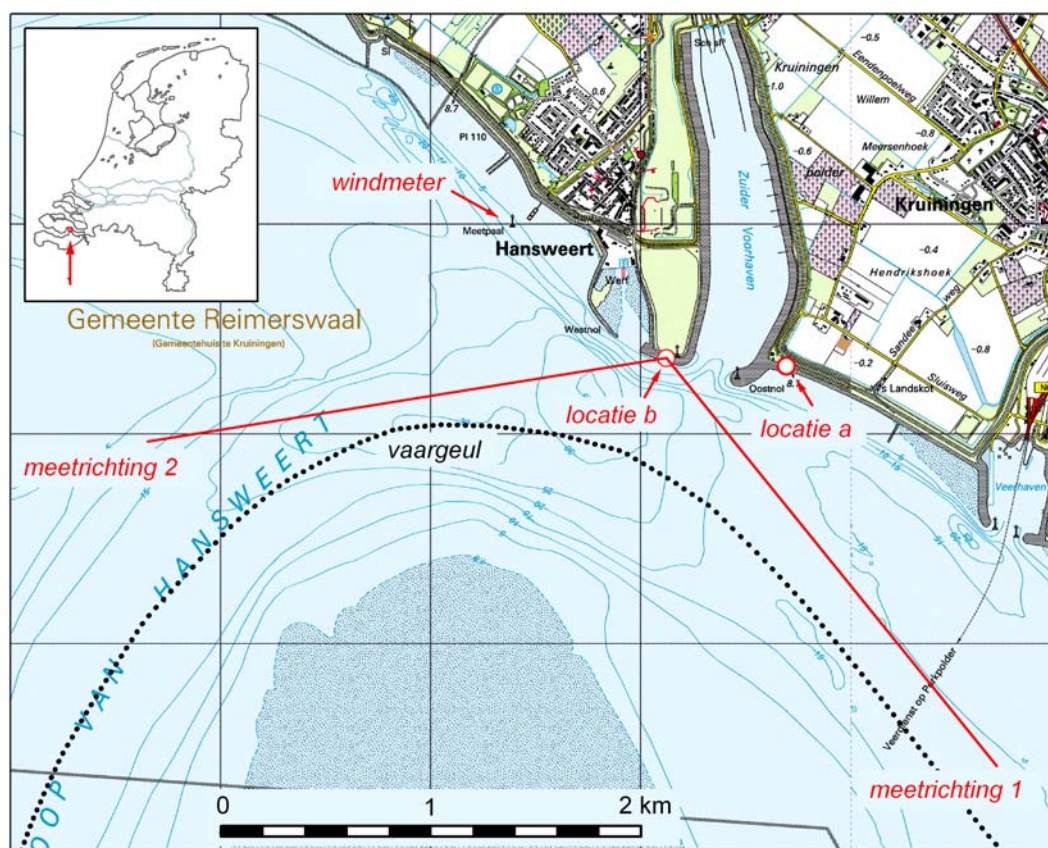


Figure 2-4. Measurement locations at Hansweert. Location a, Location b: locations where the inspection vehicle took measurements. The measurement directions are only shown for Location b. Wind gauge: measurement mast of Rijkswaterstaat³ where wind speed, wind direction and water level are measured. **Windmeter = wind gauge; meetricting=measurement direction; locatie=location**

The measurements discussed in this report were all conducted on seagoing vessels on the Westerscheldt. Hansweert was chosen as a measurement location because the shipping channel is near the coast and because the scanning plane can be located both parallel to the direction of travel and perpendicular to the most likely wind directions. See Figure 2-4 for an overview of the measurement location, and Figure 2-5 for a photograph of the inspection vehicle at location b.

Initially, location a was chosen, next to the Hansweert radar post. In May 2006, this location was used to take measurements. In June and October 2006, the inspection vehicle was moved to location b; this was because large-scale construction activities were taking place on the dike and breakwater at location a. During the measurements, there was periodic communication with the radar post.

³ Directorate for Public Works and Water Management



Figure 2-5. The inspection vehicle at the measurement location.

An automated wind gauge is located near this measurement location; this wind gauge is part of the ZEGE measurement network (*Zeeuwse getijdenwateren*). This measurement network is maintained by the Hydro Meteo Centrum Zeeland (HMCZ), a sub-department of the Rijkswaterstaat Zeeland Directorate. The wind and tidal data are published on the Internet (www.hmcz.nl) and were used to calculate the emission factors in the present report. The wind speed was calculated at the elevation at which the lidar measurement indicated that the smoke plume was present; this was done by using a logarithmic wind profile, the measured wind speed and the measured water height.

On a measurement day, the following procedure was used. Upon arrival at the location, the inspection vehicle was first stabilized and levelled. The orientation of the vehicle with respect to the north was then determined. After this, based on the dominant wind direction on that day, a measurement direction was chosen. The laser and the telescope were then calibrated to each other for every angle of inclination. At this point, the system was ready to take measurements of a passing ship.

For every passing ship, the following procedure was used. The instrument began taking measurements when the ship approached, but was not yet within measurement range. From this point on, complete scans of the vertical plane were made continuously. At a certain point, the wind blew the smoke plume of the ship through the measurement plane, which could be seen from the measurement signals. The smoke plumes were visible in a sequence of scanning plane measurements. Measurements continued until the smoke plume of the ship could no longer be seen in the measurement signals.

The measurements were processed by determining the concentration at various locations in the plume, and then multiplying this concentration with the corresponding plume area and the wind speed at that elevation. After this, all partial contributions were added up across the entire plume surface. In this way, an emission factor was determined for every scanning plane measurement. Because the smoke plumes of all ships were visible in a sequence of scanning plane measurements, more than one emission factor could be determined for all ships. In this way it could be determined how the emission developed during the period of approximately 5 minutes when the plumes of most ships were visible.

2.4 Determining an emission factor from a measurement



Figure 2-6. The HMS Rotterdam, shortly before passing the inspection vehicle.

To show how the emission factor was determined, the measurements conducted on the HMS Rotterdam (Figure 2-6) will be used as an example. This ship sailed through the Westerscheldt on 9 October 2006. On that day, due to the direction of the wind, the lidar was aimed to the southeast (measurement direction 1 in Figure 2-4). At approximately 10:30 hours UTC⁴, the smoke plume of this ship entered the scanning plane of the lidar. The SO₂ concentrations that were measured at that time are shown in Figure 2-6. In this figure, the horizontal axis shows the distance to the lidar, and the vertical axis shows the elevation above the water surface. Note that the vertical axis is extended with respect to the horizontal axis; in reality the scanning plane is much more elongated than is shown in the figure. The colour of the plane indicates the concentration of SO₂.

⁴All times in this report are given in UTC (*Universal Time Coordinated*). However, UTC is two hours behind Central European Summer Time (CEST), which was the local time in the Netherlands during the research; 10:30 hours UTC is therefore 12:30 hours local time.

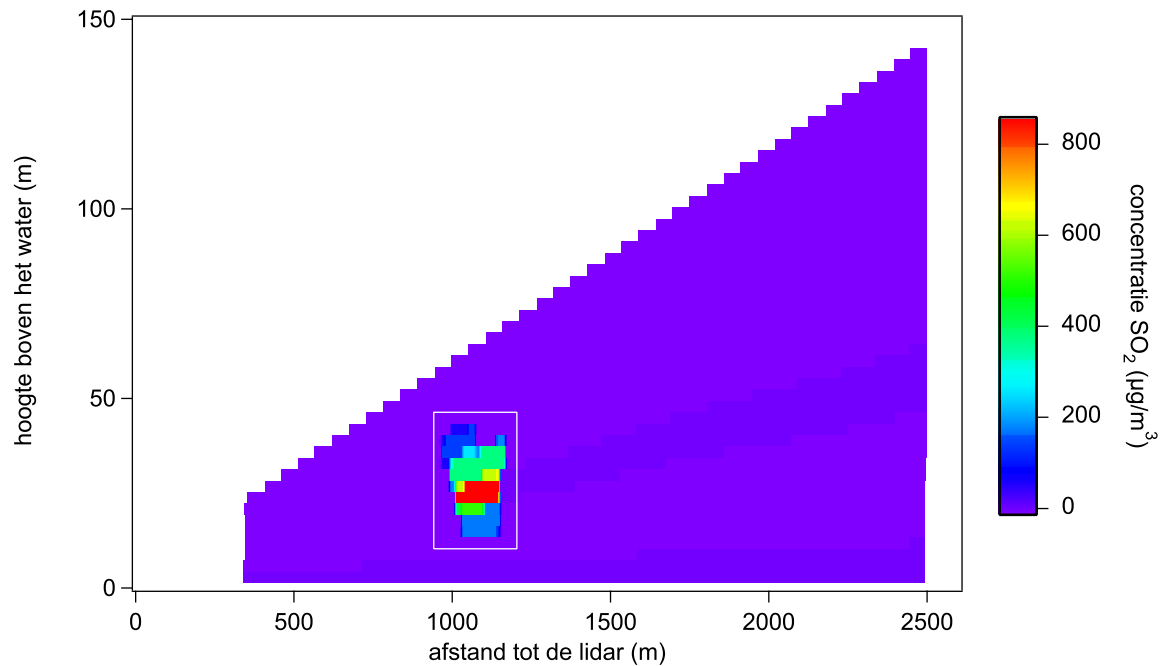


Figure 2-6. Cross section through the smoke plume of the HMS Rotterdam. the colour indicates the concentration of SO₂ in the air. The white rectangle shows the plume as it was used in the further analysis. **Hoogte boven het water = elevation above the water; concentratie = concentration; afstand tot de lidar = distance to the lidar**

To process this data into an emission factor, in Figure 2-6 the plume has been selected (the white rectangle in Figure 2-6; this selected area is shown in Figure 2-7 A). For every elevation, the total quantity of SO₂ at that elevation is determined. This results in a gas load curve (also shown in Figure 2-7 A). By multiplying this by the wind profile (Figure 2-7 B), corrected for the angle between the wind direction and the scanning plane, and then adding up all values, the emission factor can be found. For this ship at that time, the emission factor was 7.1 g per second.

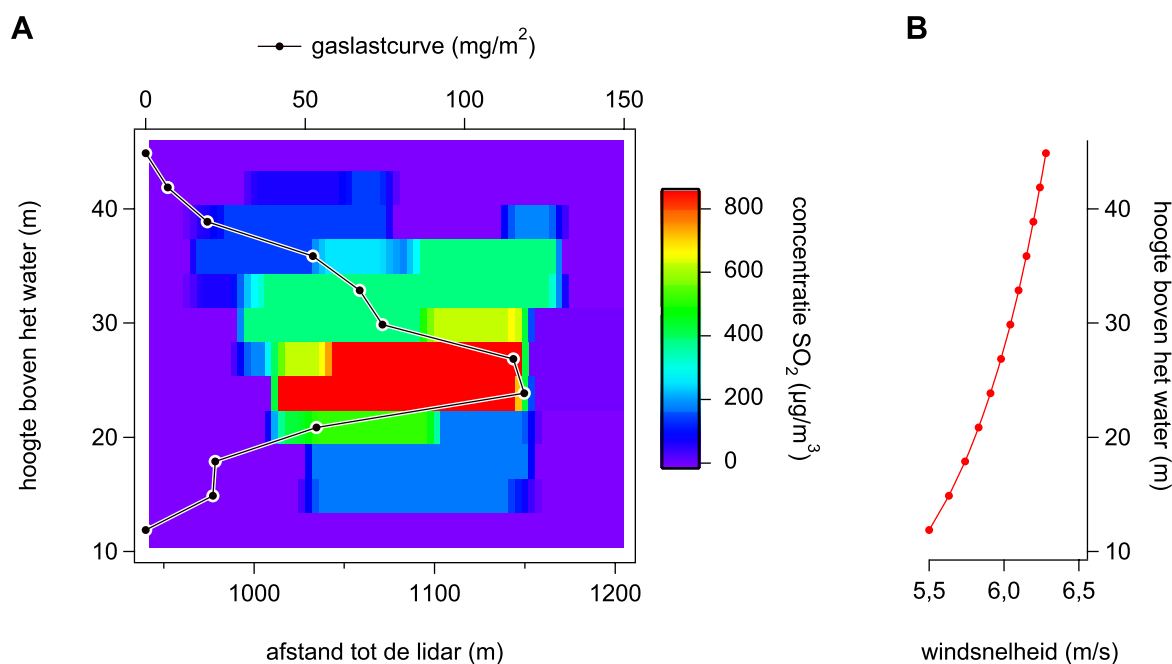


Figure 2-7. **A:** cross-section from Figure 2-6 of the smoke plume of the HMS Rotterdam, and the corresponding gas load curve. **B:** logarithmic wind profile.

Gaslastcurve=gas load curve; **hoogte boven het water** =elevation above the water;
concentratie=concentration; **afstand tot de lidar** = distance to the lidar;
windsnelheid= wind speed

As stated in Section 2.3, the wind speed used in the calculations was measured at the nearby wind mast of Rijkswaterstaat. Every 10 minutes, this measurement mast generates data for wind speed and wind direction, among other things. It also measures the water level. The wind speed used for the calculations is the velocity measured at the mast reduced to the velocity at 10 m above sea level. The logarithmic wind profile is calculated from the wind speed and the water level (Figure 2-7 B). Figure 2-8 shows the wind and water data as measured by Rijkswaterstaat on 9 October 2006, with all ships measured on that day. From this data, a wind speed, wind direction and water level can be determined for every ship at the time it passed the measurement location. Because a passage takes less than 10 minutes (a ship remains within range of the lidar for no more than five minutes), a single emission factor for each passage is sufficient, even though multiple emission factors per ship were determined for each passage.

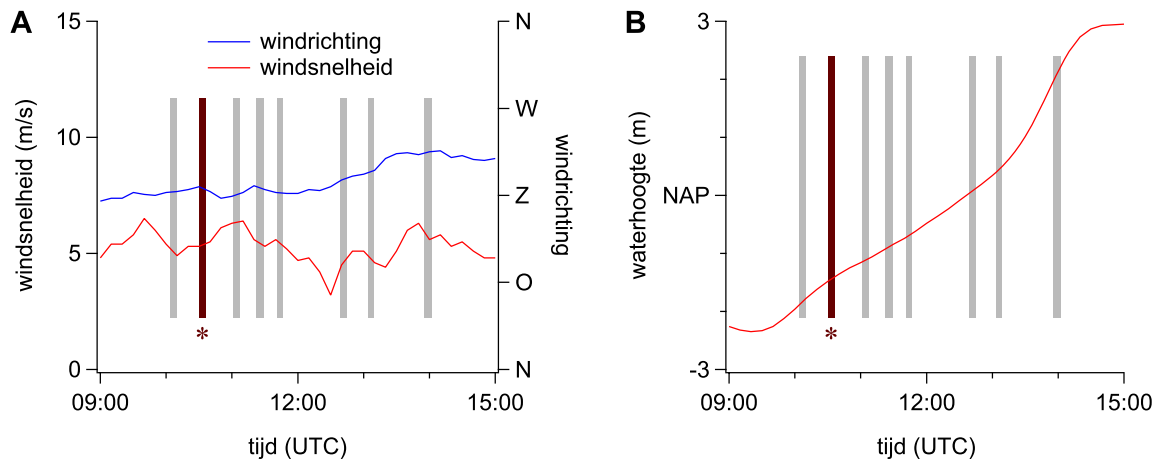


Figure 2-8. Wind and water data at Hansweert, measured by Rijkswaterstaat, on 9 October 2006. The ships measured on the state are shown with grey bars. The ship discussed in this example, the HMS Rotterdam, is marked with a *. **A**: wind speed, reduced to 10 m elevation, and wind direction. **B**: water level. **Windsnelheid = wind speed; wind richting = wind direction; water hoogte = water level**

In Figure 2-6, the plume can be clearly distinguished from the background. It is also clear that the entire plume is in the picture. However, during the measurement days there were situations where this was not the case. For example, it regularly happened that the plume was located so close to the beginning of the scanning plane that part of the plume was not yet in the picture. In those cases, however, the entire plume was usually in the picture during the previous or subsequent scanning plane measurement, so that an emission factor could still be determined. It also happened that two ships passed each other just as their smoke plumes came into the picture. In that case, the smoke plumes could not be distinguished from each other and no emission factor could be determined.

3 Results

Sections 3.1 - 3.5 briefly describe the conditions on each measurement day and report the emission factors for all ships measured on that day. Section 3.6 discusses the results of a determination of the lower limit of quantification. Finally, the results are summarized in Section 3.7.

3.1 Measurement results on 16 May 2006



Figure 3-1. One of the ships measured on this day, the Probo Emu, as it passed the inspection vehicle.

On this day, the inspection vehicle was set up at location a (Figure 2-4). The average wind speed was 3.3 m per second; the wind direction was southwest to west-northwest (220 to 279°⁵). The measurements were conducted towards the southeast (measurement direction 1 in Figure 2-4). The water level varied from -1.18 m to 0.13 m with respect to NAP. The temperature was 16 °C, cloudy, but no precipitation.

Measurements were conducted on the smoke plumes of 7 ships. Two or more emission factors could be allocated to 5 of the ships. The results are shown in Table 3-1.

⁵ Degrees east of north. A wind direction of 270° is therefore a westerly wind.

Table 3-1. Results of emission measurements on 16 May 2006.

- a In: sailing towards Antwerp. Out: sailing towards Vlissingen.
 b The time interval that the plume of the ship was visible on lidar. Times are given in UTC (see note 4, page 6).

name of ship	in/out^a	time (UTC)^b	emission (g/s)
MSC Jade	In	11:48-11:50	10
			9.8
			21
Probo Emu	In	12:00-12:02	23
			48
			33
Blexen	Out	12:36-12:41	1.8
			1.5
			3.5
			2.8
			2.6
			2.6
Arklow Rainbow	Out	13:13-13:16	4.9
			2.3
			1.1
			2.9
			0.91
Chopin	Out	13:23-13:26	1.7
			2.8
<i>JA Sunrise</i>	<i>Out</i>	<i>13:26-13:35</i>	<i>plume too close to plume of Stolt Inspiration, analysis impossible</i>
<i>Stolt Inspiration</i>	<i>In</i>	<i>13:26-13:35</i>	<i>plume too close to plume of JA Sunrise, analysis impossible</i>

3.2 Measurement results on 21 June 2006



Figure 3-2. The Tai Shan as it sailed past the inspection vehicle.

On this day, the inspection vehicle was set up at location b. The wind was strong, 9.8 m per second on average; the wind direction was southwest (209 to 213°). The measurements were conducted towards the southeast (measurement direction 1). The water level varied from -1.63 m to 0.44 m with respect to NAP. The temperature was 19 °C, cloudy, with occasional showers.

Measurements were conducted on the smoke plumes of 5 ships. Two or more emission factors could be allocated to all these ships. The results are shown in Table 3-2.

Table 3-2. Results of emission measurements on 21 June 2006.

name of ship	in/out	time (UTC)	emission (g/s)
Margareta B	out	11:49-11:51	0.64
			6.1
Maersk Malacca	out	12:04-12:08	30
			33
			45
Tai Shan	in	12:10-12:14	17
			20
			16
			19
			19
Izmir Express	out	12:46-12:49	7.9
			27
Ek-River	out	12:54-12:57	5.3
			5.4
			4.6

3.3 Measurement results on 23 June 2006

*Figure 3-3. The Vijitra Naree as it sailed past the inspection vehicle.*

On this day, the inspection vehicle was set up at location b. There was little wind, 0.8 to 1.9 m per second (average 1.4 m per second); wind direction varied from southeast to southwest (151 to 219°). Measurements were conducted towards both the southeast and the west (measurement directions 1 and 2). The water level varied from -0.63 m to 2.50 m with respect to NAP. The temperature was 20 °C, mostly clear, no precipitation.

Measurements were conducted on the smoke plumes of 11 ships. Two or more emission factors could be allocated to 3 of these ships. Emission factors could not be determined for the other 8 ships due to the lack of wind. The smoke plume of these ships was either not blown through the scanning plane at all, or this took so long that the plume could no longer be recognized as such in the lidar signal. The results are shown in Table 3-3.

Table 3-3. Results of emission measurements on 23 June 2006.

name of ship	in/out	time (UTC)	emission (g/s)
Bastiaan Broere	out	10:06-10:08	0.22 9.5
<i>Kristin Knudsen</i>	<i>in</i>	<i>10:08-10:10</i>	<i>plume did not go through scanning plane</i>
<i>Sichem Marbella</i>	<i>out</i>	<i>12:21-10:45</i>	<i>plume did not go through scanning plane</i>
<i>Trout</i>	<i>out</i>	<i>10:21-10:45</i>	<i>plume did not go through scanning plane</i>
Vijitra Naree	in	11:45-11:50	2.9 5.4 7.4 8.3 5.5 2.1 1.7
<i>Swalinge</i>	<i>in</i>	<i>11:55-12:03</i>	<i>plume did not go through scanning plane</i>
<i>MSC Eyra</i>	<i>in</i>	<i>12:08-12:10</i>	<i>plume did not go through scanning plane</i>
MSC Mee May	out	12:17-12:20	3.1 2.7 3.1 2.9 3.6
<i>Betsy S</i>	<i>in</i>	<i>12:20-12:22</i>	<i>plume did not go through scanning plane</i>
<i>Rhonestern</i>	<i>in</i>	<i>12:29-12:42</i>	<i>plume did not go through scanning plane</i>
<i>Atlantic Cartier</i>	<i>out</i>	<i>12:46-12:50</i>	<i>plume did not go through scanning plane</i>

3.4 Measurement results on 9 October 2006



Figure 3-4. The Jilihu, shortly before passing the inspection vehicle.

On this day, the inspection vehicle was set up at location b. The average wind speed was 5.2 m per second and initially came directly from the south, but later shifted to the southwest (177 to 226°). The measurements were conducted towards the southeast (measurement direction 1). The water level varied from -1.96 m to 2.14 m with respect to NAP. The temperature was 18 °C, mostly cloudy. There was no precipitation at the measurement location, although there were nearby showers around 13:00 hours UTC. Measurements were conducted on the smoke plumes of 12 ships. Three or more emission factors could be allocated to 10 of these ships. The results are shown in Table 3-4.

Table 3-4. Results of emission measurements on 9 October 2006.

name of ship	in/out	time (UTC)	emission (g/s)
MSC London	in	10:04-10:10	25
			31
			21
			26
			15
HMS Rotterdam	out	10:30-10:36	12
			14
			7.2
			7.1
			7.9
Altair	in	10:39-10:46	plume did not go through scanning plane
			2.4
MSC Maureen	out	11:01-11:07	64
			26
			23

name of ship	in/out	time (UTC)	emission (g/s)
Betsy S	out	11:22-11:29	16
			6.6
			3.5
			3.7
			1.4
			1.3
NCC Hijaz	in	11:41-11:46	15
			13
			8.6
			13
			20
Happy Girl	out	12:39-12:45	4.5
			6.7
			5.9
			5.0
<i>Neera Naree</i>	<i>in</i>	<i>12:45-12:49</i>	<i>plume did not go through scanning plane</i>
CS AV Rio Rapel	in	13:01-13:04	27
			30
			13
			18
Neveska Lady	out	13:05-13:12	26
			13
			18
			10
			16
Manzanillo II ⁶	out	13:55-13:58	1.6
			1.4
			1.1
Jilihu	in	13:58-14:02	1.6
			7.8
			0.44
			1.6

⁶ Utility ship, after passing the measurement location it worked on the concrete shore protection on the Westerscheldt.

3.5 Measurement results on 10 October 2006



Figure 3-5. The Stena Forecaster as it sailed past the inspection vehicle.

On this day, the inspection vehicle was set up at location b. The wind speed was 3.0 m per second on average; the wind direction was southeast for most of the day, but briefly before the final measurement it shifted to the east (73 to 136°). The measurements were conducted towards the west (measurement direction 2). The water level varied from -1.36 m to 2.33 m with respect to NAP. The temperature was 18 °C, mostly cloudy. During the morning, it rained briefly (from 10:00 to 10:40 hours UTC), in the afternoon there was no precipitation.

Measurements were conducted on the smoke plumes of 7 ships. Three emission factors could be allocated to only one these ships. The fact that emission factors could not be determined for the other ships was, similar to the measurements on 23 June (page 21), due to the lack of wind. The smoke plume of these ships was either not blown through the scanning plane at all, or this took so long that the plume could no longer be recognized as such in the lidar signal. The results are shown in Table 3-5.

Table 3-5. Results of emission measurements on 10 October 2006.

name of ship	in/out	time (UTC)	emission (g/s)
<i>Southern Juice</i>	<i>in</i>	<i>11.05-11.13</i>	<i>plume did not go through scanning plane</i>
<i>Manzanillo II</i>	<i>out</i>	<i>11.21-11.27</i>	<i>plume did not go through scanning plane</i>
<i>Sloman Challenger</i>	<i>in</i>	<i>11.27-11.33</i>	<i>plume did not go through scanning plane</i>
<i>MSC Marta</i>	<i>in</i>	<i>14.07-14.13</i>	<i>plume did not go through scanning plane</i>
<i>Al-Sabahia</i>	<i>in</i>	<i>14.23-14.31</i>	<i>plume did not go through scanning plane</i>
<i>Seaturbot</i>	<i>out</i>	<i>14.30-14.35</i>	<i>plume did not go through scanning plane</i>
Stena Forecaster	out	14:41-14:44	2.8 1.9 2.0

3.6 Determining the lower limit of quantification

The limit of quantification of the measurements was based on the measurement results in situations where no smoke plumes were present from ships sailing past. These measurements were used to determine an emission factor; this was done in the same way (see Section 2.4) as for the measurements where ships were present. This determination was carried out for six scanning plane measurements, all of which were performed on 9 October 2006. The emission factors are shown in Table 3-6. The average of these six emission factors provides an estimate of the lower limit of quantification: 0.1 g SO₂ per second.

Table 3-6. Results of emission measurements without smoke plumes, 9 October 2006.

time (UTC)	emission (g/s)
11:24-11:26	0.11
12:44-12:45	0.06
10:30-10:31	0.14
11:01-11:02	0.22
11:45-11:45	0.07
13:05-13:05	0.09
<i>average</i>	<i>0.1 ± 0.1</i>

3.7 Summary of all measurement results

During the five measurement days, measurements were conducted on a total of 42 ships. An emission factor could be determined for 24 ships. A summary of the measurement days is shown in Table 3-7.

Table 3-7. Summary of the measurement days.

a Measurements were conducted on the smoke plumes of this number of ships.

b An emission factor could be determined for this number of ships.

c The average wind speed on this day.

d The range of the wind directions on this day.

Date	ships measured ^a	ships with emission factors ^b	wind speed (m/s) ^c	wind direction (°) ^d
16-05-2006	7	5	3.3	220-279
21-06-2006	5	5	9.8	209-213
23-06-2006	11	3	1.4	151-219
09-10-2006	12	10	5.2	177-226
10-10-2006	7	1	3.0	73-136
<i>all days</i>	<i>42</i>	<i>24</i>		

There were three very successful measurement days – 16 May, 21 June and 9 October – during which an emission factor could be determined for 20 of the 24 measured ships. On the other two measurement days, 23 June and 10 October, emission factors could be determined

for only 4 of the 16 ships measured. Chapter 4 discusses the factors that can determine whether or not the emission of a passing ship can be measured.

Table 3-8. Results of emission measurements.

name of ship	date	number of measurements	average emission (g/s)
MSC Jade	16-05-2006	3	14 ± 6
Probo Emu	16-05-2006	3	35 ± 12
Blexen	16-05-2006	6	2.5 ± 0.7
Arklow Rainbow	16-05-2006	5	2.4 ± 1.6
Chopin	16-05-2006	2	2.2 ± 0.8
Margareta B	21-06-2006	2	3.4 ± 3.9
Maersk Malacca	21-06-2006	3	36 ± 8
Tai Shan	21-06-2006	5	18 ± 1
Izmir Express	21-06-2006	2	17 ± 13
Ek-River	21-06-2006	3	5.1 ± 0.4
Bastiaan Broere	23-06-2006	2	4.9 ± 6.6
Vijitra Naree	23-06-2006	7	4.8 ± 2.6
MSC Mee May	23-06-2006	5	3.1 ± 0.3
MSC London	09-10-2006	5	24 ± 6
HMS Rotterdam	09-10-2006	6	8.4 ± 4.2
MSC Maureen	09-10-2006	3	37 ± 23
Betsy S	09-10-2006	6	5.5 ± 5.7
NCC Hijaz	09-10-2006	5	14 ± 4
Happy Girl	09-10-2006	4	5.5 ± 1.0
CS AV Rio Rapel	09-10-2006	4	21 ± 7
Neveska Lady	09-10-2006	5	17 ± 6
Manzanillo II	09-10-2006	3	1.4 ± 0.3
Jilihu	09-10-2006	4	2.9 ± 3.3
Stena Forecaster	10-10-2006	3	2.2 ± 0.5

Table 3-8 shows an average emission factor for each ship measured, including a standard deviation. This is the average of the 2 to 7 emission factors as listed in Table 3-1 through Table 3-5. The standard deviation is an indication of the variation in the individual emission factors. The number of emission factors is also listed.

4 Discussion, conclusions and recommendations

In this chapter the most important characteristics of the lidar method are discussed. Section 4.1 analyses the factors that determine the chance of success of the lidar measurement (i.e. where the measurement provides a value for the sulphur dioxide emission). Section 4.2 discusses a number of performance characteristics of the method, such as measurement uncertainty, lower limit of quantification and selectivity. The chapter ends with the most important conclusions of the entire study.

4.1 Factors that determine the chance of success of the lidar measurement

In the study, the measurement technique resulted in an emission factor for more than half of the passing ships. The following discussion addresses the factors that determined whether or not an emission factor could be obtained with the lidar technique. At the end of the chapter, the results are summarized in a text box.

The role of wind direction

During the study, the inspection vehicle usually operated from the same measurement location near Hansweert. Due to the position of the shipping route with respect to this location, it turned out that suitable measurements could be conducted only with the wind blowing from the southwest to southeast (approximately 90 degrees on the compass). It is only with these wind directions that the scanning planes can be located more or less perpendicular to the smoke plume and are also close enough to the ships to conduct the measurement. Fortunately, the wind frequently blows from these directions in the Netherlands. Nevertheless, during the study the wind direction played an important limiting role in the use of the measurement technique. This was partly due to an unfortunate coincidence: during the measurement weeks that were reserved for the study, the wind blew more than average from a different direction. This clearly showed that for operational usage it is desirable to have access to multiple locations which are suitable for various wind directions. It would then be easier to realize a planned number of measurement days during a given period..

The role of wind speed

There were five measurement days during the study. On three of these days, virtually all measurements resulted in an SO₂ emission factor. However, on the other days fewer measurements resulted in an emission factor, ranging from less than half to only a few. The essential difference appeared to be the wind speed. On days with little wind, there were a number of factors that worked against a successful SO₂ measurement:

- The plumes of the ships spread out more, resulting in a larger and more diffuse plume;
- The wind speed itself was more difficult to determine, which affected the uncertainty of the measurements;

- At low wind speeds, the wind direction was often more variable. This also affected the measurement result. Moreover, the data for a number of ships could not be used because the plume crossed the measurement plane insufficiently or not at all.

The study showed that at a minimum wind speed of 5 m per second, equivalent to 3 Beaufort, these problems no longer play a role. Certainly at the coast and offshore, lower wind speeds occur seldom. The fact that this did occur during the study was due to two reasons. Firstly, it was initially assumed that low wind speeds would be advantageous, since the concentrations would be higher. Secondly, at the end of the study less suitable measurement days had to be used due to the approaching deadline.

Other limitations

The only other serious limitation is precipitation. Rain has a negative influence on the optical echoes which lidar uses. Moreover, the measurement setup was not entirely rain proof. In the Netherlands, it rains approximately 6% of the time, and somewhat less on the coast.

Finally, it should be noted that the same inspection vehicle was also being used for other environmental measurements (see page 7). As a result, the instrument was not always available on call. However, it was possible to reserve the instrument for a specific period..

The probability of success summarized

The meteorological situation ultimately determines the probability of success. There must be sufficient wind, and the wind must blow from a suitable direction for the measurement location. The weather must also be dry. If these conditions are met, then there is a very high probability that a large number of ships can be measured per measurement day.

It is essential that a number of measurement locations are available which are suitable for various wind directions. If this condition is satisfied, most of the limitations will be eliminated.

However, it will still be impossible to guarantee beforehand that measurements can be conducted on a specific date.

4.2 Performance characteristics of lidar emission measurement

This section discusses a number of performance characteristics of the lidar emission measurement, such as measurement uncertainty, lower limit of quantification and selectivity. At the end of the section, the results are summarized in a text box.

Measurement uncertainty

Chapter 3 presented the results of the various scans for a large number of ships; these scans lasted approximately 45 seconds each. Each scan can be thought of as an independent measurement of the emission. For all ships, more than one scan could be conducted; in this way more than one emission factor could therefore be determined for all ships. These figures sometimes show a large deviation. What causes this deviation and what does this mean for the measurement uncertainty?

The role of emission variability

First of all, it should be noted that in a number of cases the measured differences can be correctly attributed to actual differences in the emission. On the Westerscheldt, the ships do not sail a straight course at an even speed. During the measurement days, a sudden change in soot emission showed a number of times that the ships were "giving full gas". The actual emission was therefore not always constant.

The measurement uncertainty of the lidar method itself

There is measurement uncertainty with the lidar method, as there is with any other method. Factors that play a role in this uncertainty include the variability of the wind (the direction and the wind speed), the meandering of the smoke plume and the measurement uncertainty of the lidar method itself. Appendix 1 addresses these aspects in greater detail.

Based on the study, it is impossible to make a statement about which of the above factors – true emission variations or measurement uncertainty – provide the largest contribution to the deviation of the results of the various scans. However, an upper limit of the uncertainty of the lidar measurement can be derived if it is assumed that the emissions of the ships do not vary at all. This analysis is shown in Appendix 1. The measurement uncertainty of the lidar measurement is approximately 20%, but this upper limit includes the emission variations of the ships. There are indications that the lidar method itself is more precise, because for a number of ships much more precise measurements were obtained.

Lower limit of quantification

As shown in Section 3.6, the lower limit of quantification was ascertained by determining an emission factor in the portion of the atmosphere where no smoke plume was present in the scanning plane. The lower limit of quantification was determined to be 0.1 g of sulphur dioxide per second. As a rule, the emissions measured in this study were significantly above this limit.

Selectivity

The selectivity of the lidar method is determined by the presence or absence of gases other than the target gas (in this case SO₂) for which the lidar is sensitive. As explained in Section 2.1, lidar is sensitive to a gas that – similar to the target gas – differentially attenuates the two colours of light that the lidar emits. The difference in attenuation determines the sensitivity. For each target gas, the colours are chosen in such a way that the sensitivity for the target gas is maximized and that for other gases is minimized. Nevertheless, it is impossible to eliminate beforehand the possibility that another gas could also differentially attenuate the colours of light used, and therefore could result in a false-positive or false-negative measurement. Such a gas would affect the sensitivity of the smoke plume measurements discussed in this report only if it is present in the smoke plume in a sufficient quantity.

The most obvious gas with a possible influence on sensitivity is nitrogen dioxide (NO₂). Interference caused by this gas has been investigated. For other gases that undoubtedly appear in the smoke plume, such as water vapour, carbon dioxide, nitrogen monoxide, carbon monoxide and hydrocarbons, spectroscopic interference is less likely. However, this possibility has not been investigated specifically.

The sensitivity for NO₂ turned out to be much lower than that for SO₂. The lidar colours used in the study are more than 400 times more sensitive for SO₂. Moreover, the sensitivity has an opposite sign: an NO₂ concentration of +428 µg/m³ is seen as an SO₂ concentration of -1 µg/m³. The nitrogen dioxide emission of the ships is definitely lower than 10 g per second; otherwise the smoke plumes would be visibly yellow. These emissions are therefore on the same order, or lower than, the sulphur dioxide emissions. Therefore, considering the 400 times greater sensitivity to SO₂, the possibility of underestimation due to the presence of NO₂ is negligible.

As part of a more extensive validation study, the selectivity could be investigated in greater detail by analyzing a typical smoke plume with conventional analytical chemistry methods, and then determining the effect on the lidar measurement of every component found. However, such a study was beyond the scope of the present research.

Summary of performance characteristics of the lidar measurement

As they sailed past the measurement location in this study, the ships could be scanned four times on average. Based on the four scans, the sulphur dioxide emission could be determined with a measurement uncertainty of approximately 20%. This value is an upper limit.

The smallest sulphur dioxide emission that the method can detect was determined to be 0.1 g per second. As a rule, the ship emissions measured in this study were significantly above this limit.

Limited research has shown that the method has few problems with other trace gases in the ship emissions. Specifically, distortion of the measurement by nitrogen dioxide has been eliminated.

4.3 Recommendations for future research

The measurement method is operational within the indicated framework and can be used as such in subsequent studies where the sulphur dioxide emission of seagoing vessels is determined with a measurement uncertainty of approximately 20%. In this regard, the technology is certainly suitable for use as a surveillance and detection instrument.

If the technology is to be used as an enforcement instrument, a more extensive validation of the measurement method as a whole would be desirable. Important aspects in this regard are ascertaining the *precision* and *accuracy* of the method itself. This study should preferably be conducted using a stationary source with a known, constant sulphur dioxide emission. As part of the study, the lidar method would be compared with other, more conventional techniques for ascertaining the emissions.

In addition, the use of this technology in an enforcement instrument will require further verification of its selectivity, although at this time strongly interfering gases are not expected to be found. The best method is the above-mentioned complete plume analysis, with a calculation of the effects on the lidar measurement. However, most components can be evaluated in a simple fashion by means of a limited literature study or a brief spectroscopic analysis.

5 Conclusions

This study has shown in practice that it is possible to make remote measurements from the shore of the sulphur dioxide emissions of seagoing vessels while they are underway. The measurement system used in this study was placed in an inspection vehicle that is entirely self-sufficient. This methodology is fully operational and is available for the VROM Inspectorate as a detection or screening technology for ships that use sulphur-rich and sulphur-poor fuels.

At this time, the instrument has a range of 2.5 km and a lower limit of quantification of 0.1 g per second. We found that the upper limit of measurement uncertainty for a typical emission determination for a single ship was approximately 20% when using the lidar technology. This upper limit of 20% also includes the variability of the ship emissions. The results from several individual ships suggest that the precision of the lidar method itself is higher, perhaps much higher.

In contrast to our expectations beforehand, the study showed that the best results were obtained at somewhat higher wind speeds (above 5 m per second, equivalent to 3 Beaufort). At these wind speeds, the precision of the measurement and its probability of success were the highest.

The most important advantage of the method is that the measurements could be conducted remotely and that the crew of the ship therefore did not realize that the measurements were taking place. A second advantage is that – unlike the conventional methods used in practice – virtually every ship that passed could be measured. This provides a major improvement in efficiency.

An important disadvantage is that only a limited number of wind directions can be measured from a single measurement location. To increase the usability of the method and to allow the measurement sessions to be scheduled more effectively, it is therefore desirable to have access to multiple measurement locations that are suitable for various wind directions.

Within current legislation, which focuses primarily on the sulphur content of the fuel, the lidar method will initially have a role as a detection and screening instrument. During this process, the instrument would be used in combination with conventional methods. Regarding enforcement, measurements conducted with the lidar method would then help to select ships to be boarded and tested with other methods.

As an independently operating enforcement instrument, the lidar method is presently usable for ascertaining those exceedences where estimated fuel consumption is sufficient to demonstrate that fuel with an excessive sulphur content is being used. In view of the large differences in sulphur content between fuels that are permitted and forbidden, this application

appears to be realistic. Due to the absence of data about fuel consumption in this study, however, the above statement cannot be supported in greater detail.

The lidar measures the actual emissions of the ship. If legislation was in place that specified not only the fuel, but also the actual emission, the lidar method could provide a better contribution to enforcement.

On land, the sulphur dioxide emissions of industrial installations are limited by means of permits. These permits are based on the Netherlands Emission Guidelines for Air (NeR, April 2003), which imposes additional demands on sources that emit more than 2 kg sulphur dioxide per hour (0.56 g per second). In all 24 cases, the measured emission of the seagoing vessels exceeded this level. The highest measured emission was 36 g per second. Attention to ocean shipping as a source of air pollution is therefore important for both making regulations and enforcing them.

References

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Acknowledgements

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As part of this study , we also used the wind measurements from the automated wind gauge at Hansweert, which are made available to the public by the Hydro Meteo Centrum Zeeland on the internet (www.hmcz.nl).

Appendix 1 Determining the precision of the emission measurement

Chapter 3 presented the results of the various scans for a large number of ships; these scans lasted approximately 45 seconds each. Each scan can be thought of as an independent measurement of the emission. For all ships, more than one scan could be conducted; in this way more than one emission factor could therefore be determined for all ships. These figures sometimes showed a large deviation. This deviation is discussed below.

The role of emission variability

First of all, it should be noted that in a number of cases, the measured differences can be correctly attributed to actual differences in the emission. On the Westerscheldt, the ships do not sail a straight course at an even speed. During the measurement days, a sudden change in soot emission showed a number of times that the ships were "giving full gas". The emission was therefore not constant.

The role of wind variability and the uncertainty of wind measurement

When measuring the emission, the determination of the wind speed plays an important role. If the wind speed is overestimated or underestimated by a specific percentage, then the estimated emission factor becomes too high or too low by the same percentage. For each scan, the local wind speed during the 45 seconds that the plume is scanned is the most important. In the present study, however, we used the data from the measurement mast operated by Rijkswaterstaat (see page 13), which is located several kilometres from the smoke plume (see also Figure 2-4). A study of the variability of this wind data shows that this distance results in an uncertainty in the emission factor of approximately 10%. The variability in the wind direction plays a much smaller role, as long as the plume is located more or less perpendicular to the scanning plane.

The role of smoke plume meandering

During the lidar scan, which lasts approximately 45 seconds, the laser beam moves in nine steps from the bottom to the top of the scanning plane. At every measurement direction, the beam remains still for five seconds and measures the concentration distribution in that direction. The entire scanning plane is therefore not measured simultaneously, but is scanned from the bottom to the top. However, during these 45 seconds, the plume also moves; frequently the plume axis moves both up and down and from left to right and back again. This movement is called meandering and can be seen very clearly in Figure 3-4. Due to this meandering, it is possible that the plume in the scanning plane could coincidentally move with the scan (from bottom to top), or could move opposite to the scan (from top to the bottom). In the first case, the plume would remain in the picture too long and an excessive emission would be measured; in the second case the measured emission would be too low. The magnitude of this effect is difficult to quantify, but could be significant for individual

scans if the meanders are large. However, it is clear that this effect quickly averages out in sequential scans. With a strong wind, the meanders are smaller.

The role of measurement uncertainty in the concentration measurement

The remote measurement of concentration with lidar has its own uncertainties. Because only a brief measurement time for each measurement direction is available, the concentration measurements are based on a relatively noisy echo signal. The measurement uncertainty of the ascertained quantity of SO₂ in the plume is approximately 10% with heavily loaded plumes. With lightly loaded plumes, the uncertainty is larger.

Precision of the method as a whole

It is difficult to quantify the precision of the method as a whole. Traditionally, this is determined by calculating the statistical variation based on a sufficient number of sufficiently accurate measurements repeated on the same sample. This was not possible in the present study because the sample was always different. Nevertheless it appears to be possible to determine an upper limit of the precision based on the present study. We made this determination as follows:

- (1) We ascertained that the above-named factors that play a role in the variability virtually all lead to a *relative* variation in the result, regardless of the source strength itself. If every ship could be normalized with its true source strength, then we could consider all ships to be statistically identical. The only real exception to the above is variability caused by human actions ("giving full gas").
- (2) For every ship, the average emission was determined based on all successful scans (Table 3-8). Each individual scan was then normalized using this average. After this, each scan was expressed as a percentage of the average for the corresponding ship, where the sum of all averages was 100%.
- (3) After this, the clearest cases of "giving full gas" were eliminated. These are the ships where one or several scans strongly deviated from the others. All data from these ships were eliminated from additional analysis. This concerned 2 of the 24 ships in the study.
- (4) Finally, the statistical variation of the total set of remaining scans was analyzed. This concerned 86 scans of 22 ships.

The results are shown in Figure B-1.

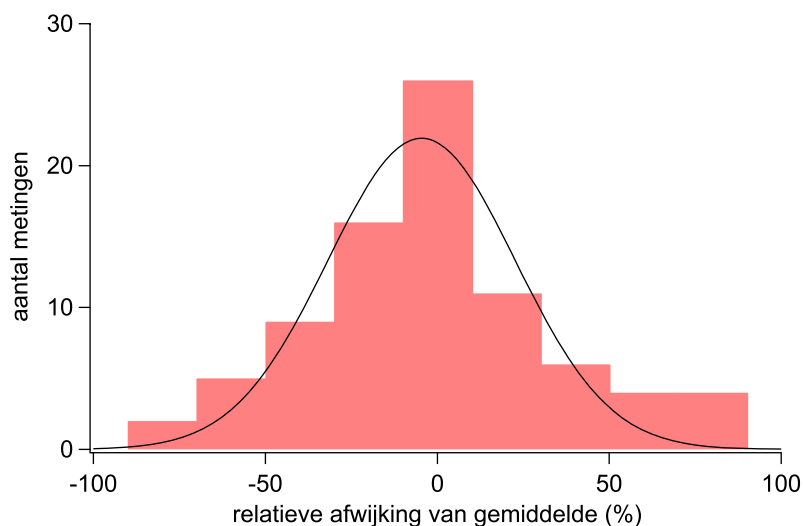


Figure B-1. The histogram shows the distribution of the results of individual scans, expressed as a percentage of the average result for the corresponding ship. In black, a Gausse curve has been fitted to the histogram. The deviation is 38% (1 sigma).

Aantal meetingen = number of measurements; relatieve afwijking van gemiddelde = relative deviation from mean

It can be concluded that the individual scans have a 1-sigma deviation of 38%. A typical emission factor for a ship, as shown in Table 3-8, has been calculated as the average of four scans and has a precision of 19% (1 sigma).

It should be noted that 19% is an estimate for the upper limit of the precision of the lidar method based on virtually all successful scans in the study. It is an upper limit because the observed deviation has been calculated including the variation in source strength (the change in actual emission from scan to scan), and it is unknown how large this variation is. It is very possible that the lidar measurement itself is significantly more precise and that the observed deviation is determined primarily by variations in the emission. The significantly better results from some individual ships appear to point in this direction (Tai Shan: 18 ± 1 g per second, compared to MSC London: 24 ± 6 g per second, in both cases five scans and a comparable emission). To determine the precision of the lidar measurement itself, repeated measurements of a source with constant strength are required. This was not a part of the pilot study, but has been proposed as part of a follow-up study.