

Transport Research Arena (TRA) Conference

Assessing Shipping Induced Emissions Impact on Air Quality with Various Techniques: Initial Results of the SCIPPER project

Sokratis Mamarikas^a, Volker Matthias^b, Matthias Karl^b, Lea Fink^b, Pauli Simonen^c, Jorma Keskinen^c, Miikka Dal Maso^c, Erik Fridell^d, Jana Moldanova^d, Åsa Hallquist^d, Johan Mellqvist^e, Vladimir Conde^e, Ruud Verbeek^f, Jan Duyzer^f, Daniëlle van Dinther^f, Hilka Timonen^g, Jukka-Pekka Jalkanen^g, Anu-Maija Sundström^g, Elisa Majamäki^g, Antonios Stylogiannis^h, Vasilis Ntziachristos^h, Tim Smythⁱ, Mingxi Yangⁱ, Anthony Deakin^j, Richard Proud^j, Johannes Oeffner^k, Jonathan Weisheit^k, Jörg Beecken^l, Andreas Weigelt^l, Simone Griesel^l, Hannes Schoppmann^l, Sonia Oppo^m, Alexandre Armengaud^m, Barbara D'Annaⁿ, Brice Temine-Rousselⁿ, Grazia-Maria Lanzafameⁿ, Bettina Knudsen^o, Jon Knudsen^o, Maria Kuosa^p, Matti Irjala^p, Leo Buckers^q, Jasper van Vliet^q, Leonidas Ntziachristos^{a*}

^aAUTH, ^bHEREON, ^cTAU, ^dIVL, ^eCHALMERS, ^fTNO, ^gFMI, ^hHMGU, ⁱPML, ^jEE, ^kCML, ^lBSH, ^mATMOSUD, ⁿAMU-CNRS, ^oEXPLICIT, ^pAEROMON, ^qILT

Abstract:

This paper presents the methods deployed by the Horizon 2020 SCIPPER project to characterize emission performance of vessels, mainly under the perspective of checking compliance to new emissions regulations. Various on-board and remote measurement techniques have been demonstrated within five experimental campaigns conducted at Europe's main sea areas and ports. Almost a thousand of ship plumes has been measured and cross checked with various instrumentation, revealing the emission profile of ships during actual operation. Accuracy of each measurement technique was also tested. Emission measurements are further exploited to assess the impact of shipping on air quality of coastal areas, by identifying the transformations of pollutants performed in the atmosphere as plume evolves and quantifying onshore pollutants concentrations attributed to shipping activity.

© 2023 The Authors. Published by ELSEVIER B.V.

This is an open access article under the CC BY-NC-ND license (<https://creativecommons.org/licenses/by-nc-nd/4.0>)

Peer-review under responsibility of the scientific committee of the Transport Research Arena (TRA) Conference

Keywords *shipping emissions; emission measurements; on board and remote measurements; air quality*

* Corresponding author. Tel.: +30 2310 996003.
E-mail address: leon@auth.gr

1. Introduction

The shipping sector is still associated with relatively high quantities of harmful pollutants emitted (NO_x , SO_x , PM) despite the steps that have been made since 1997 to reduce its overall environmental footprint (Matthias et al 2010, Aulinger, et al.2016). Recent IMO and EU provisions for even lower emission limits oblige ship owners and operators to take more decisive actions than those existed in previous decades. Therefore, fuels with reduced sulphur content, alternative fuels and aftertreatment devices are main measures adopted by international and domestic navigation operators to comply with the increasingly stricter emissions regulations of the IMO and EU (Lehtoranta et al. 2019).

Although realistic emission limits and effective mitigation measures are the main elements for achieving the targets set in a well-designed environmental strategy, still these cannot stand alone. It is also critical for authorities to ensure that ships' operations are in compliance with established environmental requirements in different waters, by developing and maintaining mechanisms that can systematically monitor emission performance. In this context, the Horizon 2020 SCIPPER project (Shipping Contributions to Inland Pollution Push for the Enforcement of Regulations) deploys state-of-art and next-generation measurement techniques to monitor emissions of vessels under their regular operations. SCIPPER aims at providing evidence on the performance and capacity of different techniques for shipping emissions monitoring, to serve regulations enforcement. By collecting emissions information, the project also assesses the impacts of shipping operations to air quality, under different emission compliance scenarios. The methodology applied by SCIPPER relies on a combination of real-world emission measurements and air quality simulation activities, at different scales. All vessel types are considered by SCIPPER, including containers, Ro-Ro, tankers etc.

This paper describes the emission measurement campaigns conducted within SCIPPER and presents initial findings of the emissions characterization performed. It also analyses the process for the further exploitation of experimental results in the evaluation of shipping impacts on the air quality of EU coastal and harbour areas.

2. Measurement Campaigns

The five experimental campaigns of SCIPPER take place at main sea areas of Europe, namely the Western Baltic Sea, the English Channel, and the ports of Marseille in the Mediterranean and Hamburg in the North Sea. These locations are characterized by particularly intense shipping activity, practically of all types of vessels throughout the year, with occasional seasonality peaks, especially at the port of Marseille. They serve the bulk of maritime cargo transit within Europe, and significant numbers of ferries and cruise ships used by passengers for transportation purposes and vacations.

2.1. Overview of methods deployed in SCIPPER

During SCIPPER campaigns, pollutant concentrations have been measured in a significant number of ship plumes, with the use of various techniques (on-board, remote sniffer & optical) and many types of sensors (both high-end and low-cost). The experimental targets of the campaigns can be summarized in three main points: 1) to intercompare the various measurement techniques; 2) to characterize the emission performance of ships and develop emission factors, and 3) to assess phenomena of plume ageing and dispersion, which are critical for air quality evaluations.

One of the methods used is on-board emission measurements, using high-end equipment placed in the ship funnel. Gas analyzers, similar with those widely used in road vehicles during portable emission measurements, constitute the basis for gaseous pollutants characterization on-board. Low-cost sensor boxes also sampled the same exhaust, and their accuracy was checked in relation to the findings of the high-end systems. The intention by using such sensors is to identify their potential for long-term emission measurement on board operational vessels. For PM, a dilution system is required, and particle counters, collection filters and an oxidative flow reactor are utilized for defining particle mass and number along with chemical speciation and size under different ageing conditions. One ferry has been measured by SCIPPER with on board measurements, as is later explained.

SCIPPER also performs remote measurements using sniffers and optical techniques. Sniffer systems are placed at fixed stations mainly in the entrance of ports, downwind of passing ships. Sniffers are also installed on patrol boats and on drones that chase specific vessels from an angle that allow placing the sniffer in the plume of the target vessel for the necessary sampling time. Optical remote techniques are also installed at fixed positions similarly to stationary

sniffers and their beam crosses the target plume, whilst the potential of monitoring shipping emissions from satellites is also explored using the Tropospheric Monitoring Instrument (TROPOMI). All experimental processes applied within SCIPPER are schematically presented in Fig 1.

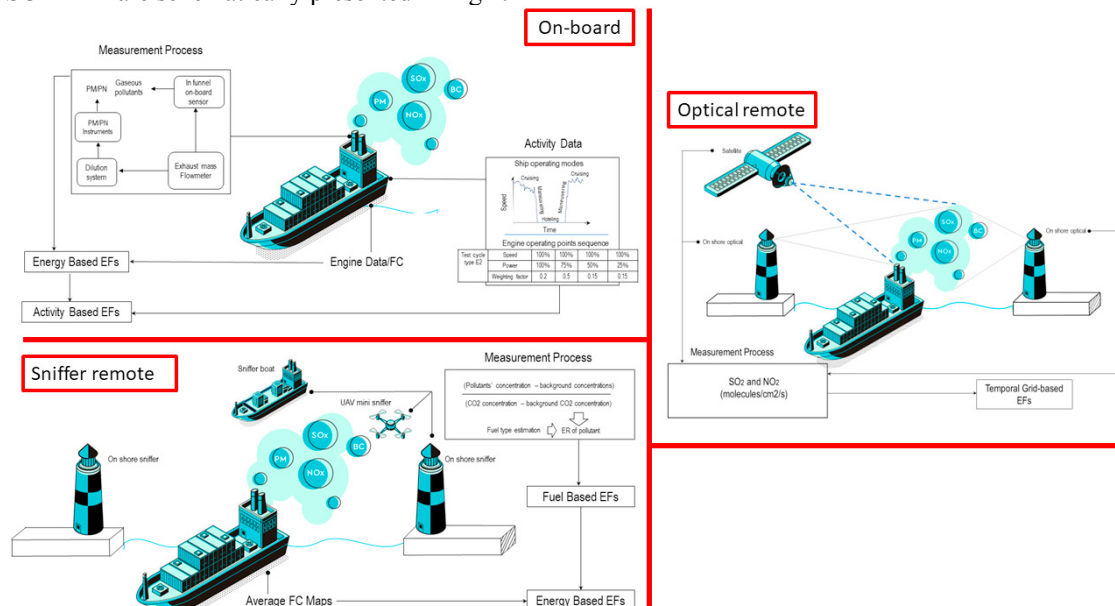


Fig. 1. Methods deployed in SCIPPER during real-world emission measurement campaigns SCIPPER (2021b); Grigoriadis et al. (2021).

This schematic representation, apart from visualizing the techniques deployed per measurement type, also describes the process for further exploitation of emission measurements in developing emission factors (EFs) for ships. The production of EFs mostly requires the identification of fuel properties and/or consumption rates, together with the measurement of pollutants' concentration in the plume. During on-board measurements, fuel consumption of the ship is measured or estimated, together with the instantaneous engine power and finally emission rates are composed by dividing the total mass of pollutants produced over fuel consumption, or the useful work delivered to the shaft. Energy based emission factors (g/kg or g/MJ fuel) can be based on pollutants' concentrations and fuel properties only considering CO₂ emissions concentrations and the carbon balance method, assuming default values for the fuel used. Also, specific emissions in g/kWh can be calculated based on pollutants' concentrations and an approximation of the specific fuel consumption (Verbeek et al., 2021, SCIPPER, 2021a). In sniffer-based remote measurements, gaseous pollutants concentrations in ppm are measured, and referring them per unit of fuel consumed is done (again) considering CO₂ emissions concentrations and the carbon balance method, assuming default values for the fuel used SCIPPER (2019). In such a process, background concentrations should be subtracted, and the exact method of correction and uncertainty estimate is a matter of research within the project. Apart from emissions quantification and the development of EFs, similar measurement techniques are used for identifying plume ageing, atmospheric transformations, and pollutants dispersion. These all provide inputs for the air quality assessments of SCIPPER.

After the overview of the methods deployed generally in SCIPPER, this paper now on describes each campaign in more detail. Also, initial indicative results are highlighted, not only concerning the emission performance of ships related to the regulated pollutants of the sector (SO_x, NO_x), but also for PM, THC and CO.

2.2. Measurements in Marseille

Two measurement campaigns conducted at the port of Marseille in France mainly captured the emission status of ships just before (July 2019) and after (July 2021) the enforcement of the 2020 global sulphur cap in marine fuels.

The first campaign was implemented in September 2019. A harbour vessel was equipped with sniffers to sample plumes of berthing and moving ships in the port. A flying drone, carrying mini sniffers, performed remote measurements on the same plumes. Additionally, air quality stations placed at Marseille port measured atmospheric

concentrations of pollutants. Results of the campaign intended to provide a) an initial evaluation regarding the compliance of ships with sulphur emissions regulations, b) a comparative assessment of remote techniques (sniffer on a boat vs mini sniffers carried by drones), including uncertainty characterization, as well as a first assessment of remote and drones' accuracy, and c) an input to the identification of the effect on air quality of shipping emissions, before the regulation of the fuel sulphur content. During the campaign, 30 ship plumes were measured by the sniffer vessel and 21 with the drone, from which 17 common ones were further exploited for intercomparing the performance of the two techniques.

The second campaign took place in July 2021. It came more than a year after the global application of sulphur cap, providing the opportunity to check compliance rates of ships under the new regulations. Within this campaign, remote techniques (sniffer vessel and drones) were again deployed and intercompared, similarly to the campaign in 2019. However, the harbour vessel was equipped with additional equipment for assessing atmospheric transformation of pollutants while the plume disperses. Moreover, pollutants concentrations were extensively measured by onshore air quality stations to identify how plumes evolve as reach the harbour and the city. In total, 126 plumes derived from 38 different vessels were gathered and analysed for accomplishing the experimental targets of the campaign.

Table 1 presents the percentage Fuel Sulphur Content (FSC) detected by the drone while making in-plume measurements during the first and second phase of experiments. Results show that no ship exceeded the 0,5% limit after the cap application, while almost half of the plumes measured were found to come from ships that operated on fuels of high sulphur content before the enforcement. This reduction of sulphur quantities in fuels is evidently followed by a respective mitigation of SO_x induced emissions.

Table 1. Plumes % FSC detected by the drone carrying a mini sniffer.

Campaign / date	Plumes measured	Plumes exceeded 0,5% FSC
Marseille (2019)	21	10 (48%)
Marseille (2021)	13	0 (0%)

The particular effect of atmospheric transformation on the chemical composition of the gas and condensed phase of ship emissions as well as the potential for forming secondary aerosol mass was studied in the campaign of 2021 using oxidation flow reactors (OFRs). In those devices, the emissions are oxidised by OH radicals in the presence of UV light, simulating atmospheric aging in an accelerated fashion. The TSAR (Simonen et al., 2017) was used in the sniffer boat measurements and the Go:PAM (Watne et al., 2018) was deployed in the stationary ship plume measurements in the harbour. For the majority of the ship plumes measured in the harbour, the particle mass after oxidation (i.e., PM_{AGED}) was significantly (more than 3 times) higher compared to the non-oxidised emission (i.e., PM_{FRESH}) as indicatively shown in Fig. 2. This suggests that ship emissions contain a substantial fraction of aerosol precursors. Hence, their contribution to air quality by source apportionment and modelling works need to consider such significant transformations

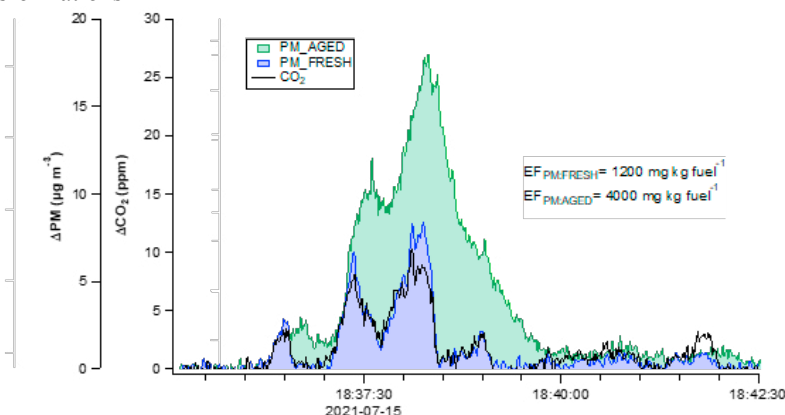


Fig. 2. Example of a ship plume measured in Marseille harbour utilising the Go:PAM OFR. PM_{AGED} and PM_{FRESH} are the particle mass concentrations measured with and without oxidation of the emissions in the OFR.

Moreover, this suggests that the contribution of vessels to total PM concentrations in the atmosphere may be higher than the concentrations calculated by emission inventorying – largely based on emission factors of primary PM – would predict.

2.3. Measurements on a ship ferry in a Western Baltic Sea route

Continuous on-board measurements on a ferry ship commuting between Gothenburg, Sweden and Kiel, Germany revealed the actual instantaneous emission profile of a large ship under various phases of its normal operation. The impact of two fuel types (MGO, methanol) and aftertreatment technology (with, w/o SCR) were also studied. Measurements took place during a period of 7 days in September 2021. The ferry ship was constantly measured while covering the trip distance of almost 400 km and emissions were characterized by high-end measurement systems placed in the ship exhaust (as well as with sensor systems). Fig. 3 presents the outputs of this characterization for various pollutants (NO_x , SO_2 , CO, THC, PM) at the 70% engine load with the use of MGO. The chart additionally specifies the effect that SCR operation and methanol use have on NO_x emissions.

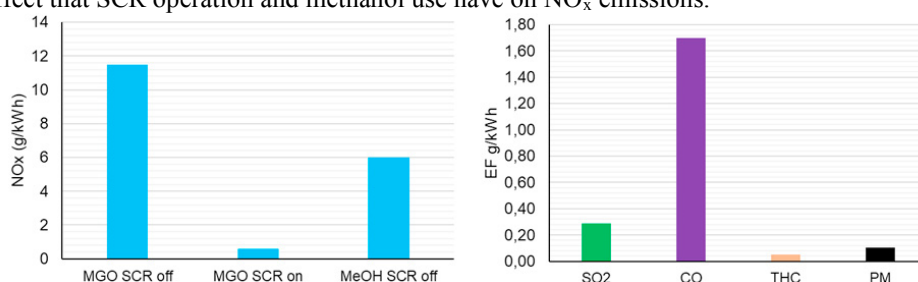


Fig. 3. NO_x emissions performance for SCR operation and methanol use (left) & EFs of various pollutants under MGO operation (right).

Experimental findings show that NO_x emissions are reduced by half with the combustion of methanol, while an almost 95% efficiency in NO_x reduction is observed during the SCR activation when the ship operates on MGO. All the pollutant emission values detected and presented in panels of Fig. 3 are rather typical of the engine technology that is found installed on such ferries for propulsion and auxiliaries' operation.

Apart from the recognition of the emission performance of this specific vessel, the campaign also targeted to check the sensitivity of the various techniques while measuring emission. Low-cost sensors conducted measurements directly in the exhaust funnel, for testing their accuracy in relation to the high-end systems. Remote techniques (sniffers, mini sniffers on drones, optical) findings were also compared with the high-end instrument readings. Remote instrumentation was placed either on the ship deck – monitoring the exhaust that exit the funnel – or at fixed onshore points where the plume reached after the ship passed by. The drones carrying mini sniffers were also departed from the same onshore points. Additionally, detailed chemical composition and physical properties of gases and particles in the exhaust were also identified. These measurements were conducted by mobile laboratories that were placed on-board, sampling from the exhaust. In particular, the effect of atmospheric aging on the composition of ship exhaust aerosol and on the formation of secondary aerosol was studied with a Potential Aerosol Mass (PAM) OFR chamber (Aerodyne Research Inc.). Fig. 4 shows that the aging of the exhaust generates secondary aerosol mass, but the secondary aerosol formation is significantly smaller with the methanol fuel than with the MGO fuel.

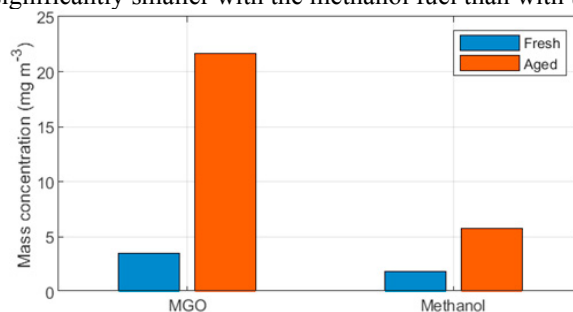


Fig. 4. Fresh and aged particle mass concentrations for MGO and methanol fuels

Within the campaign, the real-time transmission of emission measurements from the vessel to shore using binary messaging via satellite ASM (Application Specific Messaging) was also demonstrated, with excellent reporting rates. ASM is part of the VHF Data Exchange (VDE) evolution of AIS and provides a low-cost / low bandwidth communications method for reporting sensor data from remote maritime assets.

2.4. Measurements at the port of Hamburg

Remote measurements with different monitoring systems at the entrance of the Hamburg port provided a large and diversified dataset of plume measurements from various ship types, which have been also cross-checked with the different instrumentation and also by taking fuel samples for intercomparison. During the measurement campaign in Hamburg, 60 fuel samples onboard 34 different vessels were taken by the waterway police which were later analysed for their individual fuel sulphur contents (FSC) in the BSH laboratory. These vessels were measured on 34 different occasions by one or more of the remote FSC monitoring systems. The results in Fig. 5 present comparison of the results of the remotely measured FSC values with the laboratory analysed results. In general, the standard sniffers as well as the highly sensitive laser systems and experimental drone measurements tended to slightly underestimate the FSC obtained from the fuel sample, taken from the main engine. The sulphur contents of the laboratory analysed fuel samples for the comparison to the remote measurements were found to be between 0.03 and 0.11 %S m/m with a median at 0.08 % S m/m. The campaign in Hamburg accumulated in total 966 plume measurements from 436 different vessels, which will be further exploited for comparing the remote techniques, not only regarding the sulphur content detection, but also for NO_x monitoring and other pollutants identification. Detailed analysis of measured NO_x and particle emissions is ongoing.

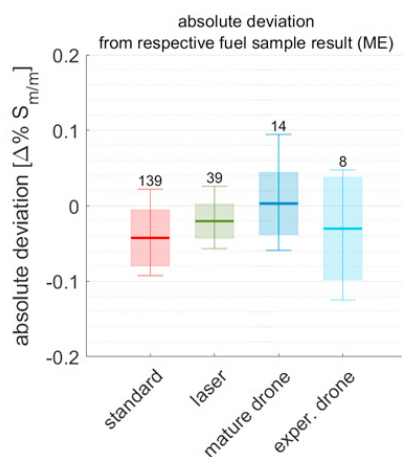


Fig. 5. Comparison of fuel sulphur contents measured by remote techniques with laboratory analysed fuel samples. Number of measurements per method is indicated by the number on top of each bar. Different systems are grouped according to their underlying measurement principles.

Solid line gives mean value while thin coloured box indicates standard deviation and whiskers the 5% and 95% percentile, respectively.

2.5. Measurements in the English Channel

Previous analysis concentrated on the experimental campaigns that have already been implemented at the period where this paper is written. A fifth campaign will take place in May 2022 in the Western English Channel and within it, SCIPPER will examine the satellite observations potential for single or group of ships monitoring. Moreover, drone-based sensors vs remote sensing techniques will be again tested. Real-time data transmission through satellite ASM will be also demonstrated. So far, an environmental observatory operated in the area providing us with pollutants concentration measurements on a constant basis. Results are provided in Fig.6 in terms of SO₂ concentration monitoring timeseries from the particular monitoring station. Additionally, on board measurements have already been conducted on an experimental vessel, where the emission data occurred from of the operation of low-cost sensors have been transmitted via satellite ASM over an extended period of time – again, with excellent reporting (detection) rates.

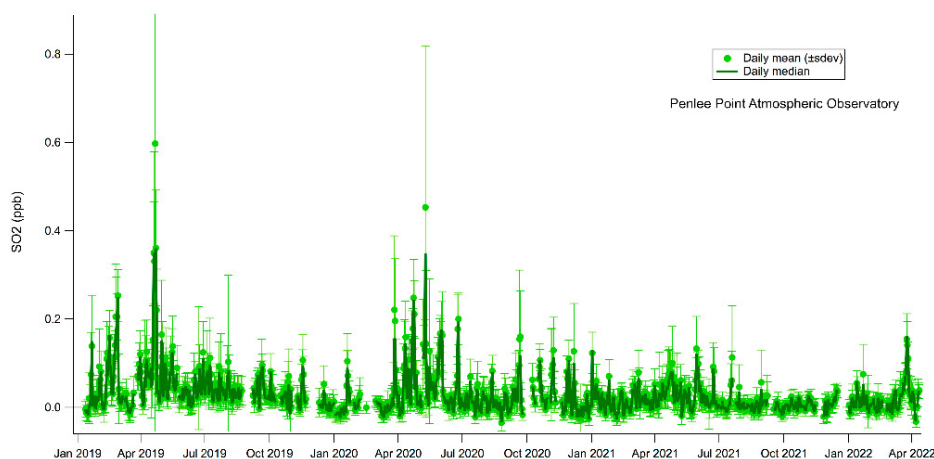


Fig. 6. SO₂ concentrations derived from an atmospheric observatory looking at the English Channel.

The potential of satellites to identify plume emissions of singular vessels has also been pre-assessed. So far, findings for NO_x show that single ship plumes are visible in TROPOMI observations (Georgoulis et al., 2020). Advanced processing allows individual plume quantification at lengths of 150-200 meters. More investigation is needed for exploiting satellite observations for compliance monitoring, but global monitoring seems to be feasible for NO₂. Other gases and aerosols detection is more challenging. Recent updates on TROPOMI SO₂ retrieval algorithms are not sufficient to bring emissions in shipping lanes in view. SO₂ signal from shipping seems to be below the detection limit of TROPOMI. Regardless of the very high spatial resolution of aerosol optical depth observations from satellites, the interpretation for shipping signals remains challenging e.g. due to other aerosol sources at the sea areas (sea salt, long range transport of aerosols).

An overview of the five SCIPPER campaigns is provided in Table 2, displaying pollutants covered, the methods deployed by SCIPPER partners, the instrumentation used and outputs regarding the number of vessel plumes captured.

Table 2. SCIPPER measurement campaigns overview

Measurement Campaign	Pollutants covered	Main methods deployed (by SCIPPER partners)	Instrumentation applied	Ships/plumes measured
Marseille (2019)	SO ₂ , CO ₂ , NO _x , BC, PN, PM ₁ , PM _{2.5} , PM ₁₀	Sniffer on vessel (Chalmers) Drone mini sniffer (Aeromon) On shore AQ (AMU, ATMOSUD)	1 boat-based sniffer, 1 UAV born mini sniffer	30 plumes
Port of Hamburg (2020)	SO ₂ , NO, NO ₂ , PM Soot	Remote sniffers (BSH, TNO, Chalmers) Optical remote (Chalmers) Mini sniffer drone (Explicit, Chalmers) Fuel sampling (BSH)	5 sniffers, 4 particle size classifiers 2 aethalometers, 1 long path DOAS 1 zenith sky DOAS, 2 UAV born mini sniffers, 1 condensation particle counter	436 vessels 966 plumes
Ferry ship Western Baltic Sea (2021)	SO ₂ , NO, NO ₂ , PM, BC, CO ₂ , NH ₃	On board characterization-High-end reference instruments and low-cost sensors (TNO, IVL, FMI, AUTH, HMGU, AEROMON, TAU, CML) Sniffer on board (Chalmers) Optical on board (Chalmers) AQ on-board (AMU, TAU, FMI, IVL) Sniffer on shore (BSH, Chalmers) Mini-sniffer drone (Explicit) Data transmission (eEE, PML)	2 high-end reference gas analysers, 5 sensor boxes, 1 optoacoustic black carbon sensor, 3 sample conditioners (diluters), 6 particle counters and classifiers Nephelometer, 2 soot monitors, 3 mass spectrometers 1 chemical reactor (aging), thermodenuder, volatility chamber, Absorbent tubes and cartridges for sampling of org. species, TEM grid samplers, 10 gas analysers 1 UAV born mini sniffer, Sniffers	1 vessel 7 days of full operation
Marseille (2021)	SO ₂ , NO _x , CO, CO ₂ , CH ₄ , PM ₁ , PM _{2.5} , PM ₁₀	Sniffer on vessel (Chalmers) Drone mini sniffer (Aeromon) AQ on a vessel (TAU, AMU) AQ on shore (AMU, ATMOSUD, IVL, TAU)	2 particles counters, 5 classifiers, 2 aethalometers, 7 sniffers, 1 FTIR VOCs, 4 mass spectrometers, 1 Xray for metals, 2 chemical reactors (study aging), 2 UAV born mini sniffers	38 vessels 126 plumes
English Chanel (Exp. vessel part)	NO _x , CO ₂	On board characterization (TNO) Data transmission (PML, eEE)	1 sensor box	21 trips

3. Contribution to air quality impact assessment

The target of air quality assessments of SCIPPER is to estimate current ship-induced air pollution footprint and predict the impact of various degrees of emissions regulations compliance at major port areas in EU. Emphasis is given on the scale of the air quality models applied and on the experimental input given from the campaigns to the advanced plume dispersion and chemical transport models (CTM) developed and applied. The STEAM model (Jalkanen et al., 2012) is applied to generate highly resolved shipping emissions (up to resolutions of 250 x 250 m²) for SO₂, NO_x, CO, VOCs, BC, OC, sulphate, ash and other PM_{2.5}. Five different regional CTMs (CMAQ, EMEP, LOTOS-EUROS, Chimere and CAMx) and two urban CTMs (uEMEP and EPISODE-CityChem) are then applied for the Mediterranean and the North/Baltic Sea area as well as for the harbour cities Marseille, Hamburg and Gothenburg.

4. Conclusions

The successful execution of the campaigns so far has led to the following main conclusions:

- It is demonstrated that distant monitoring of in-plume pollutant concentrations is technically feasible, and a variety of techniques are currently commercially available to support enforcement of regulations.
- Actual monitoring of vessels gives credible results which are on par with results of fuel sampling on board with regard to fuel sulphur levels. Such monitoring techniques seem to be feasible in satisfactorily informing enforcing authorities.
- Distant monitoring can be extended to pollutants beyond SO_x, including NO_x and particles. For all pollutants, agreements on processing, background corrections, quality assurance and exceedance thresholds are required.
- On-board measurements offer the capacity for detailed and accurate emissions characterisation. The project demonstrated the potential of low-cost sensors for on-board monitoring for SO_x, NO_x and PM (including black carbon and PN), but longer duration durability tests are required to prove their capacity for lifetime emissions.
- Real-time retrieval of on-board emissions measurements for immediate shore-based analysis has been successfully demonstrated.

Acknowledgements

The SCIPPER project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement Nr.814893.

References

- Matthias, V., et al.: "The contribution of ship emissions to air pollution in the North Sea regions." *Environmental Pollution* 158.6 (2010): 2241-2250.
- Aulinger, et al.: "The impact of shipping emissions on air pollution in the greater North Sea region—Part 1: Current emissions and concentrations." *Atmospheric Chemistry and Physics* 16.2 (2016): 739-758.
- Lehtoranta, K., et al.: "Particulate mass and nonvolatile particle number emissions from marine engines using low-sulfur fuels, natural gas, or scrubbers." *Environmental science & technology* 53.6 (2019): 3315-3322.
- Ntziachristos, L. et al.: Monitoring of ship emissions to enforce environmental regulations. The SCIPPER project. 24th Transport and Air Pollution conference, 2021.
- Verbeek, R. et al.: 'Real Sailing Emissions' for maritime vessels using sensors and satellite data transmission. 24th Transport and Air Pollution conference, 2021.
- SCIPPER, 2019: Review of available remote systems for ship emission measurements, available in scipper-project.eu, accessed April 2021.
- SCIPPER, 2021a: Assessment of on-board sensor implementation and recommendations for upgrade of most promising systems, available in scipper-project.eu, accessed April 2021.
- SCIPPER, 2021b: New set of emission factors and activity information, available in scipper-project.eu, accessed April 2021.
- Grigoriadis, A. et al.: Development of exhaust emission factors for vessels: A review and meta-analysis of available data. *Atmospheric Environment: X* 12, 100142, 2021.
- Georgoulas, A.K. et al: Detection of NO₂ pollution plumes from individual ships with the TROPOMI/S5P satellite sensor. *Environmental Research Letters* 15(12), 124037, 2020.
- Jalkanen, J-P., et al: "Extension of an assessment model of ship traffic exhaust emissions for particulate matter and carbon monoxide." *Atmospheric Chemistry and Physics* 12.5 (2012): 2641-2659.