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Performance of a remote sensing device based on a spectroscopic approach for the remote measurement of vehicle emissions

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Abstract

Today´s air pollution poses a serious health risk to human health. Much of this pollution in cities is due to vehicle emissions, hence the need to reduce them. With this purpose, a new remote sensing instrument, called 'LRSD' and developed under the EU-funded H2020 NEMO project, is presented. This newly developed system is designed to individually measure free circulating vehicles' tailpipe emissions in real-time and non-intrusively. The instrument is based on ICLs and QCLs technology, for the precise and highly sensitive detection of several pollutants (CO, CO_2, NO, NO_2, C_3H_8 and NH_3) from moving vehicles' exhaust plumes. It is designed to cover portable and fixed operation modes, as well as single or multi-lane measurements. Initial experiments show a minimum measurement accuracy of 95.41% with 10.00% deviation, with detection limits consistent with ultra-low vehicle emissions.

Keywords: Remote sensing; Emissions; TDLS; Infrared spectroscopy; ICL; QCL;

Nomenclature

RSD	Remote Sensing Device
LRSD	Laser-based Remote Sensing Device
QCL	Quantum Cascade Laser
ICL	Interband Cascade Laser
NDIR	Non-Dispersive Infrared
TDLS	Tunable Diode Laser Sensor

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1. Introduction

Air quality has become a major concern in recent years due to the negative effects of different pollutants on public health, leading to a plethora of problems, such as cardiac diseases or cancer, incurring in thousands of premature deaths per year (Manisalidis I, 2020). A large part of this pollution in cities comes from vehicle emissions, and that is the reason why these emission sources must be monitored, regulated and reduced. For instance, according to data from the inventory of polluting emissions to the atmosphere in the municipality of Madrid (Landrigan P.J, 2017), road traffic is responsible, in average, of 55% $PM_{2.5}$ air pollution and 48% NO_x air pollution in all the region, while in some areas of the city the road traffic is responsible for more than 80% of all NO_x air pollution.

The H2020 NEMO project, financed with European funds (ref. 860441), aims to reduce emissions and noise generated by traffic, thanks to the remote measurement and control of individual vehicles' noise and gaseous emissions. To provide a large-scale, cost-effective solution, the project aims to integrate novelty autonomous remote sensing systems into existing infrastructure. The new systems must be able to identify noisy and polluting vehicles for a selective control. In this direction, Urban Vehicle Access Regulations (UVAR) seek to regulate the circulation of traffic according to the emission levels of each vehicle. To do this, we must go beyond just identifying extreme vehicles (the cleanest and dirtiest vehicles), being able to differentiate small emission levels between each vehicle from remote sensors. Moreover, as new regulations impose stringent emission limits, both in type-approval and in-service (e.g. expected Euro 7), vehicle emissions tend to be lower, but at the same time there is a need to monitor that they remain low under all real-world conditions. Because of all this, the new instrument, called 'LRSD' for Laser-based Remote Sensing Device, developed and presented here, must ensure highly accurate measurements of even low concentration emissions from the circulating fleet.

The Opus LRSD is conceived to have both measurement and operative capabilities superior to currently available technologies. In particular, the system is designed to measure, within challenging specifications, lower pollutant detection limits, while keeping a stable and autonomous operation. The system is based on TDLAS technology. The first laboratory prototype presented here already shows significant improvements in the uncertainty and detection ranges of all pollutants.

2. State of the art

Spectroscopy applied to remote sensing of vehicles emissions has been studied for several years. The first RSD was developed in the late 1970s. This first unit used a technology called FTIR, based on the Fourier transform infrared spectroscopy, although it was capable of detecting different gases, it was uncommercial, unintuitive and un-autonomous (Giechaskiel, B, 2021). The following system was developed at the University of Denver (named FEAT) and patented in the 1980s, using a liquid nitrogen cooled Non-Dispersive Infrared (NDIR) sensors where the column density of each individual pollutant is measured using a broadband emission light source and individual spectral filters are tuned at each main absorption feature. This approach records the overall wavelength band light intensity, providing an averaged intensity value over the wavelength band. This method has been successfully employed to measure gaseous pollutants in the past (Carsten G. et al., 2019). The latest commercial RSD, the Opus AccuScan RSD5500, also uses NDIR technology and detects vehicles emissions using IR/UV absorption spectroscopy techniques, to accurately measure all pollutants present in the exhaust plume of motor vehicles. The equipment is equipped with a light source, which continuously emits a beam of IR/UV light at specific wavelengths for absorption of CO, CO_2, HC, NO, and NO_2 (Bishop B.A et al.,1996).

The main challenge with NDIR technology lies in the detection limit. The measurement uncertainty expands when the concentrations of pollutants present in the vehicle exhaust plume are very low. This poses a challenge in the accurate characterization of very low emitting vehicles, which would make it complicated to set individual fees based on the actual real-driving emission levels in low-emitting vehicles.

The new Opus LRSD aims to solve all these limitations. The objective is to lower the detection limit of gas concentrations under a 10% uncertainty. On the other hand, the system must be able to accurately measure NH_3, a new emerging pollutant in vehicle emissions. Vehicular NH_3 emissions are co-emitted with nitrogen oxides (NO_x) and may have a more effective pathway to particle formation in urban environments, compared to NH_3 from agricultural activities (Farren et al. 2020). From an operating point of view, the system must offer both portable and fixed installation options, it also must eliminate the necessity of equipment calibrations, it must guarantee a 24/7 operation and offer different deployment possibilities in a wide range of different environments.

3. Instrument description

The methodology used in the LRSD for measuring the gaseous species (CO, CO_2, NO, NO_2, C_3H_8 and NH_3) concentrations is tunable diode laser absorption spectroscopy (TDLAS). This is executed with 1 Interband Cascade Laser (ICL) and 4 Quantum Cascade Lasers (QCLs) installed in the main system module. The fact that the exact spectral position of the exciting light source is known, enables the measurement system to accurately draw up the absorption feature in function of the wavelength, hence significantly reducing the noise of the system and reducing the pollutants detection limits.

The system is composed of 5 main modules:

- The radiation emission module contains the 5 lasers operating in the medium-infrared spectrum. One deuterium ultraviolet lamp emits in the ultraviolet (UV) region for plume opacity measurement, as a proxy of particulate matter (PM) emission measurement.
- The detection module contains a mercury-cadmium-telluride (MCT) infrared detector and a photodiode UV detector, both synchronized with the emission module.
- The data processing module is used for real-time processing of the instantaneous absorbances of each species measured over time.
- The equipment also has a module to manage other peripherals, such as a sensor for the remote measurement of vehicle speed and acceleration, a weather station to measure the ambient conditions and a camera that records the license plates of the measured vehicles to relate them to the emissions record and later retrieve associated vehicle technical information from vehicle registry databases.

Spectroscopic remote analysis of vehicle emissions is done by calculating the ratios of pollutants to CO_2. As all gases dilute along the exhaust plume of the vehicle, the ratio of each pollutant to CO_2 remains constant along the plume (that is, NO/CO_2, NO_2/CO_2, HC/CO_2 and NH_3/CO_2). The underlying principles are detailed and reviewed in the relevant literature (Bishop, B.A., 1996), and has been applied for decades of vehicle emissions remote sensing. When performing a measurement for a vehicle passage, the system sequentially activates each of the 5 lasers to emit 5000 measurement profiles in 0.5 seconds. In other words, the device takes 1.5 million measurements in 0.5 seconds. The system takes both a “pre-car” (ambient) record and a “post-car” (ambient + exhaust plume) record. The subtraction of both measurements provides a value of the plume concentrations only.

Table 1 presents the proposed measurement ranges for the LRSD, which should guarantee a measurement accuracy of 10% within these concentration ranges for each gas. These values are consistent with the real-world emission ranges of combustion engine vehicles, covering from very clean to extremely polluting vehicles.

Table 1. Opus LRSD detection limits

Gas	Lower detection limit (ppm)	Upper detection limit (ppm)
NO	100	96000
NO_2	20	3600
CO	100	60000
CO_2	4000	720000
NH_3	100	1680
HC	50	1200

Figure 1 shows some images of the prototype LRSD, which is composed of two main subsystems: the main system contains all the optics and electronics (radiation emission module, detection module, data processing module and peripherals module), while the second subsystem is a simple reflector formed by a corner cube mirror. Light is emitted by the main subsystem, bounces off the reflector and returns back to the detector inside the main subsystem.

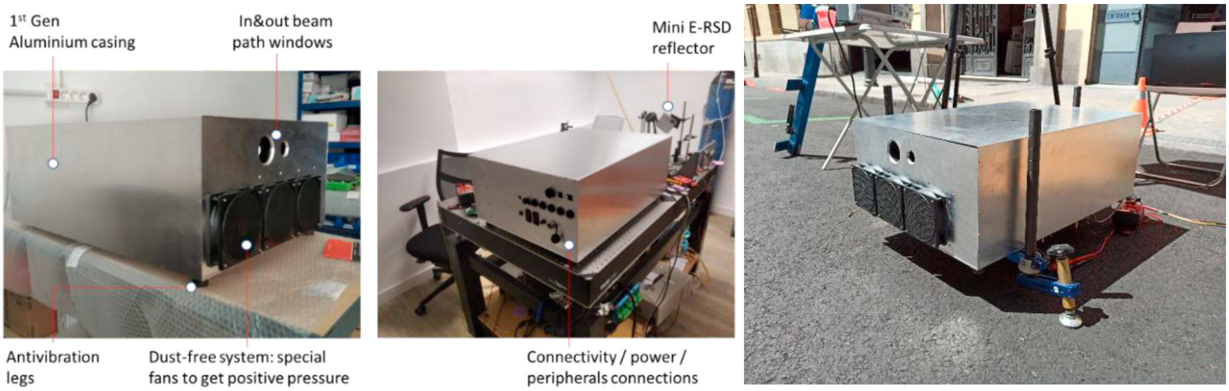

Figure 1. Opus LRSD prototype.

4. Testing methodology

To evaluate whether the first developed prototype meets the desired specifications, we have performed two types of tests: (1) laboratory tests, in a controlled, indoor environment, performing direct puff emissions with calibrated mixtures of dry gases and (2) on-road tests, in external conditions, where a vehicle with unknown tailpipe emissions is measured in real-driving conditions. We have evaluated the measurement capabilities of the LRSD for NO, NO_2, C_3H_8 and NH_3 gases against known references. Remote measurement of plume opacity has not been evaluated in this study, although the measurement method used in the LRSD is exactly the same as in legacy remote sensing devices.

4.1. Laboratory testing methodology with calibrated mixtures of dry gases

With the laboratory tests the objective is to evaluate the ability of the system to determine the relative concentrations of the different pollutants present in an open path environment. Before the tests, the absorption feature of each gas has been calibrated using several pattern gas cells. All gas cells used have dimensions of 25 mm diameter x 25 mm length and the different concentrations are shown in Table 2. (David B. Foote et al., 2021).

Table 2. Gas cells description.

Gas cell number	Composition	Concentration
1	CO_2-N_2	16% of CO_2 balanced with N_2 to 740 T
2	CO-N_2	1000ppm of CO balanced with N_2 to 740 T
3	C_3H_8-N_2	1000ppm of C_3H_8 balanced with N_2 to 740 T
4	NH_3-N_2	3400ppm of NH_3 balanced with N_2 to 740 T

Once the laser tuning control electronics is validated, and in order to validate the mathematical algorithm that calculates the gas concentrations from the measured absorbance (Parnis et al. 2013), different calibrated mixtures of dry gases are diffused in the system optical path via direct puffing. The puff release method is the standard method in the sector of vehicle emissions remote sensing (i.e. California OREMS, US EPA 420-R-96-004 and Colorado HB 1402 2001). The testing method consists in generating different dynamic column densities using an electronically controlled gas valve to diffuse gas mixtures from calibrated cylinders into the optical path of the system. With this experiment, we can validate the performance of the system for measuring dynamic gas concentrations in an open space. The calibrated gas cylinders used during the experiments are presented in Table 3. We could arrange with a certified supplier the provision of gas cylinders with lower concentrations than in the official test procedures used in remote sensing programs. The objective of this selection is to verify that the LRSD detects low concentrations of contaminants and performs precise and accurate measurements under these ranges, as the desire is that the detection limit of the equipment reaches a few particles per million.

Table 3. Calibrated gas cylinders used during the tests.

	C_3H_8 (ppm)	NH_3 (ppm)	NO_2 (ppm)	CO (ppm)	CO_2 (ppm)	NO (ppm)
Cyl_1	120 ± 0,6%	0	0	600 ± 0,5%	144600 ± 0,5 %	800± 0,7%
Cyl_2	0	0	300 ±0,5%	0	150000± 0,5%	0

Notice that NH_3 ratios could not be measured with these tests, as no bottles could be obtained with this gas including CO_2.

4.2. On-road testing methodology with a vehicle with unknown tailpipe emissions

With the on-road test the objective is to evaluate the consistency of the measurements in real-world conditions. This test was conducted in Teesdorf, Austria, where NEMO project partner Kapsch arranged for a vehicle with unknown emissions to be measured by the LRSD device during integration tests with other on-road sensing systems. The vehicle is a Suzuki Swift mil-hybrid gasoline Euro 6d utility vehicle, with nearly 40,000 km mileage, which would be representative of a vehicle with, theoretically, low tailpipe emissions, if its condition is optimal.

The LRSD prototype is placed on the side of the road, with a distance between emitter and reflector subsystems of about 8 meters, covering a 2-lane road. The test evaluates whether the remote measurement is consistent with known physicochemical principles. The vehicle is measured with an approximate speed of 50 km/h. The concentrations of the different gases present in the vehicle's exhaust plume are measured as it passes by, which are diluted over time, including turbulence and other physicochemical phenomena.

Figure 2 shows the test scheme and the measured vehicle.

Figure 2. On-road test at Teesdorf, Austria, of a Suzuki Swift mil-hybrid gasoline Euro 6d passenger car.

5. Results

This section presents the results of the tests described in previous section.

5.1. Results of the laboratory tests with calibrated mixtures of dry gases

Table 5 shows the results of the tests performed at the laboratory. Ten measurements were performed with each gas cylinder. The results show that, in the case of NO and NO_2 measurements, the system measures with an accuracy of 98.3% and 98.0% respectively, with a deviation of 4.2% and 4.0%, respectively. In the case of CO, the results show an accuracy of 95.86%, with a deviation of only 0.0004%. In the case of hydrocarbons, the accuracy is within the desired objectives, 95.4%, although slightly lower than for the other pollutants. The deviation is also higher, but only

10.0%. This means that, in the worst measured pollutant, the system is overestimating by only 5.5 ppm the real value in the measurement of the hydrocarbons.

Table 5. Results of the tests with calibrated mixtures of dry gases.

	Expected ratio (ppm/%)	Measured ratio (ppm/%)	Expected ppm	Measured ppm	Absolute difference ppm	Accuracy (%)	Deviation (%)
NO	53.33	54.26	800.00	813.90	13.90	98.30	4.24
NO_2	20.00	19.61	300.00	294.18	5.82	98.05	4.05
CO*	0.0040	0.0038	600.00	575.16	0.00	95.86	0.0004
HC	8.00	7.63	120.00	114.5	5.5	95.41	10.00

*CO ratio units are expressed as CO%/CO2%, as explained in the literature (Bishop B.A et al.,1996).

The results of the above table are plotted in Figure 3, including the individual measurements for each pollutant.

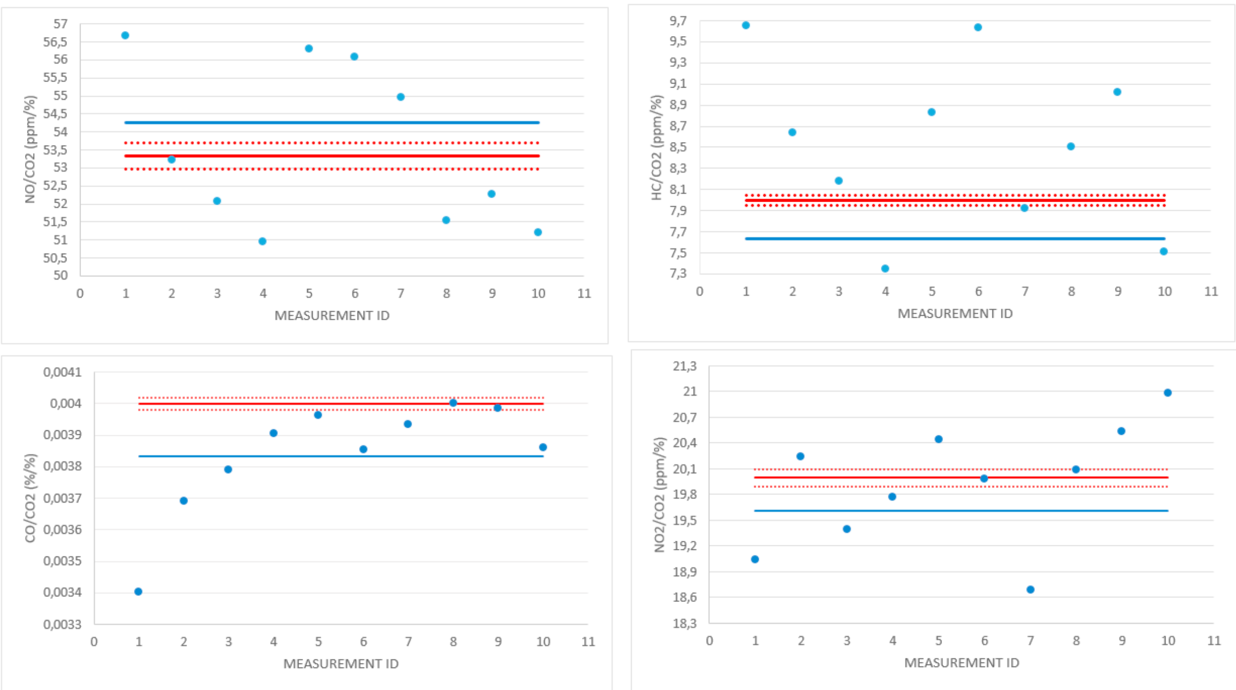

Figure 3. Results with laboratory tests with calibrated mixtures of dry gases. The x-axis represents each measurement id (a different individual test). The y-axis represents the ratio of pollutant to CO_2 concentration. Blue dots represent the calculated emission ratios from the LRSD measurements for each individual test. The horizontal dash black line represents the average emission ratio from all the LRSD measurements. The horizontal solid red line represents the average emission ratio from the gas bottle. The two horizontal dash red lines represents the ratio uncertainty from the gas bottle gas concentrations.

5.2. Results from on-road tests with a vehicle with unknown tailpipe emissions

A measurement performed with the LRSD in Teesdorf is shown in Figure 4. For each record, the system provides 32 individual data points for each gas. The data points start just after the vehicle body ends up passing in front of a sensor that activates the trigger of the measurement system, which is positioned a few centimeters away from the LRSD. The graph on the upper left (a), shows the measured data points of CO_2 column density (ppm*cm) within 0.5

seconds of a vehicle pass by. As expected, the column density first increases and then lowers with time. The CO_2 peak is detected around the 8-10th measurement data point, indicating the maximum plume concentration of the vehicle exhaust plume. From the peak, the CO_2 concentration decreases due to the dispersion in the environment. The other graphs of the figure (b, c, d, e and f) show the calculated emission ratios of NO, NO_2, CO, HC and NH_3 to CO_2. As explained in section 2, it is to be expected that these ratios remain fairly constant along the exhaust plume dilution, at least from the CO2 peak (around 8-10th data point). Indeed, it is observed that he ratios remain stable since around the 10th measurement data point. This is consistent with the expected phenomena and indicates that the first data points of a measurement may be discarded and/or that the triggering mechanism should be improved to align with the CO_2 peak. A reasonably flat plateau is observed in the calculated ratios, excluding the first data points of the plume. These results can be used to refine the measuring method.

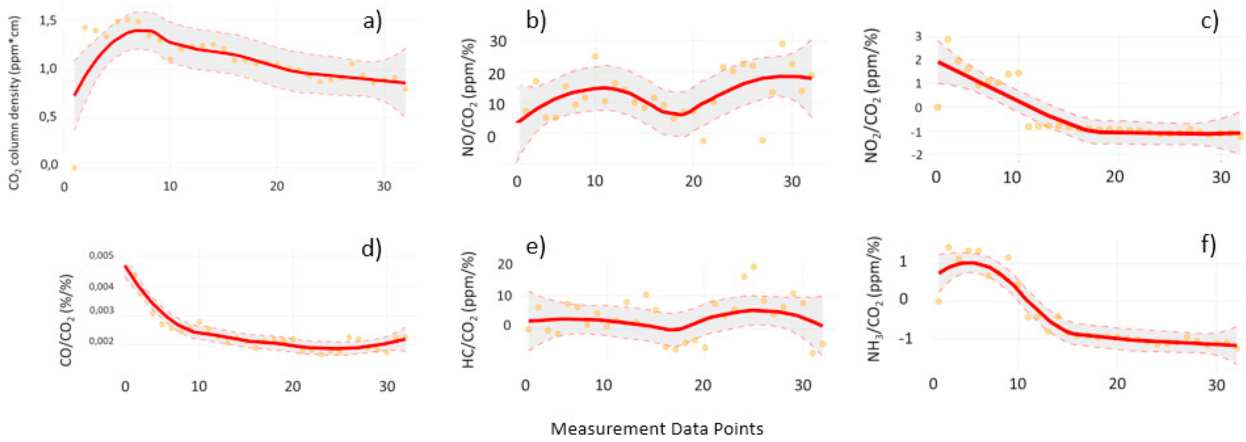

Figure 4. Distribution of measurements under real conditions of the different pollutants over time. a) CO2 column density, b) NO ratio to CO2, c) NO2 ratio to CO2, d) CO ratio to CO2, e) HC ratio to CO2 and f) NH3 ratio to CO2.

Table 6 shows the measured concentrations (average of data points 10 to 32) of this vehicle. According to the measurement, the average emissions of this vehicle are low, especially in NO_2 where 23 ppm are detected, approaching the desirable detection limit, as expected from this vehicle.

Table 6. Results of the on-road test.

	Measured ratio (ppm/%)	Measured ppm
NO	12.31	184.70
NO_2	1.54	23.17
CO*	0.002	0.036%
HC	5.06	75.84
NH_3	<6.7	<100

*CO ratio units are (CO%/CO2%)

The measurements of NH_3 are under 100 ppm, considered to be below the detection limit of the instrument. However, this is not to imply that the system cannot even improve the detection limits shown in Table 1. In fact, in general, it is observed that the dispersion of individual measurement data points from such a low-emitting car is also very low, which is an indication that the instrument might be able to measure even lower concentrations with little uncertainty.

6. Conclusions and next steps

From the experiments performed we can determine that this first version of the prototype can measure low concentrations of CO, HC, NO and NO_2 (600,120,800 and 300 ppm respectively), with an accuracy between 94% and 98.3%, with deviations of less than 10%. On the other hand, measurements of a real car are consistent with expectations and very low dispersions are observed in the calculated ratios.

Considering that this is a first non-commercial version, developed in a short time and with a prototype design, the results are very promising. The reduction of the pollutant detection limit is a breakthrough, especially for the emission characterization of the least emitting vehicles in the coming decades.

Various hardware and software enhancements may improve these specifications by a wide margin. Upgrades are already being developed, readjusting the algorithms for detecting the absorption spectra, improving the electronics to eliminate noise interfering with the measurements to lower the detection limit and modifying the design of some internal components of the system. Improved identification of the CO_2 peak in order to calculate emission ratios from this data point promises to improve averaged pollutant emission ratios.

Several validation tests are planned in the upcoming months. In May 2022, the Joint Research Centre (JRC) of the European Commission will intercompare the remote measurement of vehicles' emissions by the LRSD prototype with Portable Emissions Measurement Systems (PEMs) installed on several vehicles. Gasoline, diesel and GLP passenger cars, from Euro 3 to Euro 6 standards, will be tested. Measurement campaigns will be carried out in the cities of Madrid and Florence in the year 2022, to characterize the real-world emissions of vehicles circulating in different city locations with the LRSD. The system will also be tested to measure exhaust emissions from diesel trains in Valencia Port.

Another major improvement, in addition to the cross-road configuration presented in this document, is the top-down configuration system, under current development in the project too, to measure the emissions from vehicles circulating on multi-lane roads.

From an operating point of view, fully autonomous operation for both cross-road and top-down configurations has been confirmed, as the system's performance has been tested for hours in Teesdorf, without any detected deviations, neither required calibrations of any type. This will allow to have an RSD with portable and fixed operation capabilities, totally unassisted from human presence, with different ways of deployment and integration in European road and street environments, guaranteeing a low operational cost in long-term remote monitoring programs.

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