Are asphalt pavements an important source of atmospheric particles precursors?

J. Lasne[1], A. Lostier[1], T. Salameh[1], M.N. Romanias[1] E. Athanasopoulou[2], D. Karagiannis[2], N. Kakouri[2], S. Myriokefalitakis[2], and E. Gerasopoulos[2]

[1] Institut Mines-Télécom Nord Europe, Univ. Lille, Center for Energy and Environment, F-59000 Lille, France
[2] Institute for Environmental Research & Sustainable Development, National Observatory of Athens, Penteli, GR-15236, Greece

General context – Urban air pollution and macroscopic surfaces

Global population is growing, and increasingly concentrated in cities: **56% today, > 70% by 2050.**

- Significant and multiple sources of air pollutants
- Low natural vegetation, and high-density infrastructures (**or macroscopic surfaces**), that limit air flows, leading to pollution trapping.
- **Macroscopic surfaces account for 60-90% of the total area of an urban city**

Poor air quality related to millions of death per year.

Significant effort for air pollution reduction from major pollution sectors (Energy, solvents, transport).

New important but undocumented sources of pollutants identified such as Volatile chemical products (VCPs). VCPs account for **37 to 53 % of total VOCs in urban environments.**

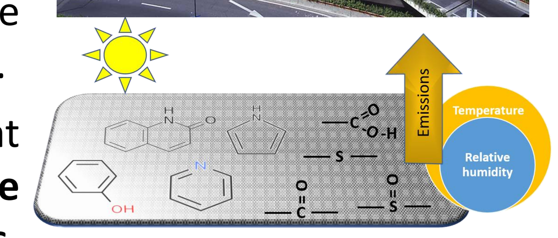
Urban air quality models often fail to reproduce NOx concentrations[1], **leading to unrealistic future predictions.**

It is urgent to better characterize the pollutants emissions sources in urban environments. What is the impact of macroscopic surfaces?

Asphalt pavements corresponds to around 40% of the area of an urban city.

Asphalt is a petroleum byproduct, composed of a large number of organic species. Temperature and humidity impact the physicochemical properties of the material and thus their emission efficiency.

Recently, Khare et al.,[2] reported that **asphalt pavements is an important source of urban pollutants and particles. However, conclusions drawn were questionable.**



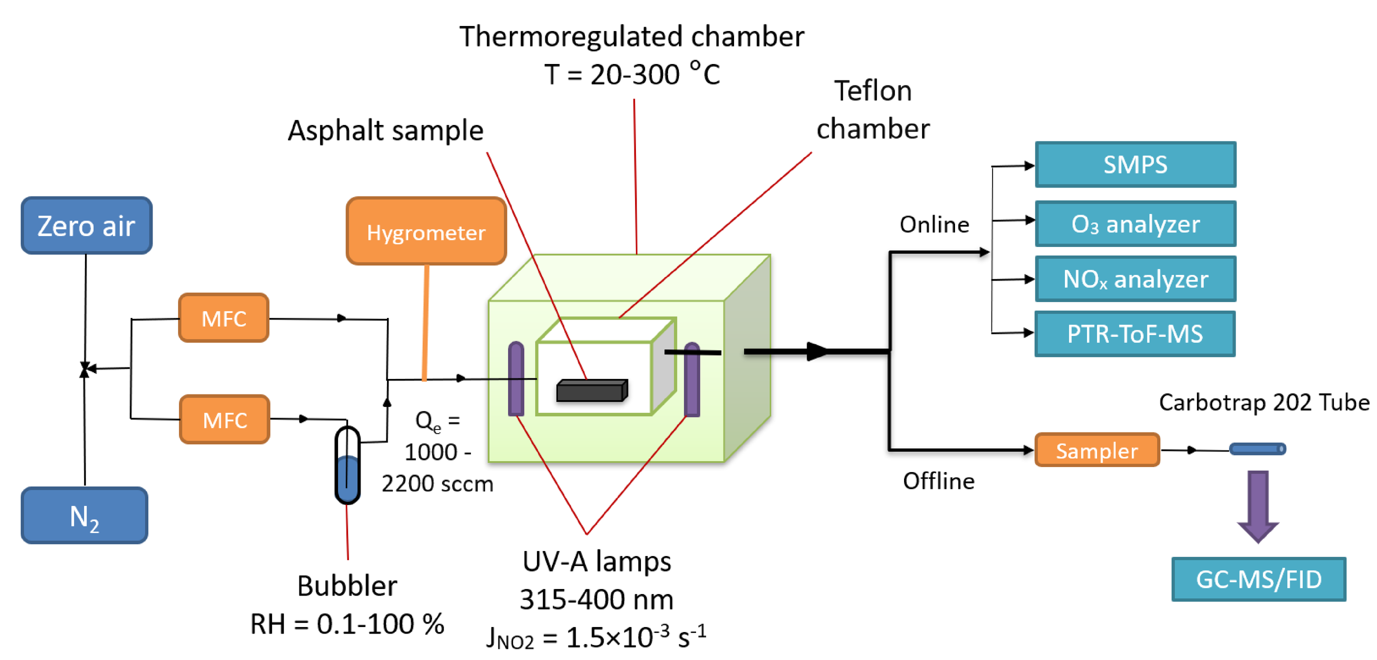
Objective of the study

Determine the emission efficiency of fresh and aged asphalt pavements	Laboratory experiments Measurement of emission factors, EF (µg m^{-2}h^{-1})
Evaluate the impact of atmospheric conditions (temperature, humidity, sunlight irradiation) on the emissions efficiency of asphalt surfaces	
Estimate the impact of asphalt surfaces on urban air quality. Evaluate the need for future mitigation strategies, and pollution control policies for asphalt pavements.	Atmospheric numerical modeling

Materials and methods

Samples Origin: Douai, North France

Thermally regulated atmospheric simulation chamber

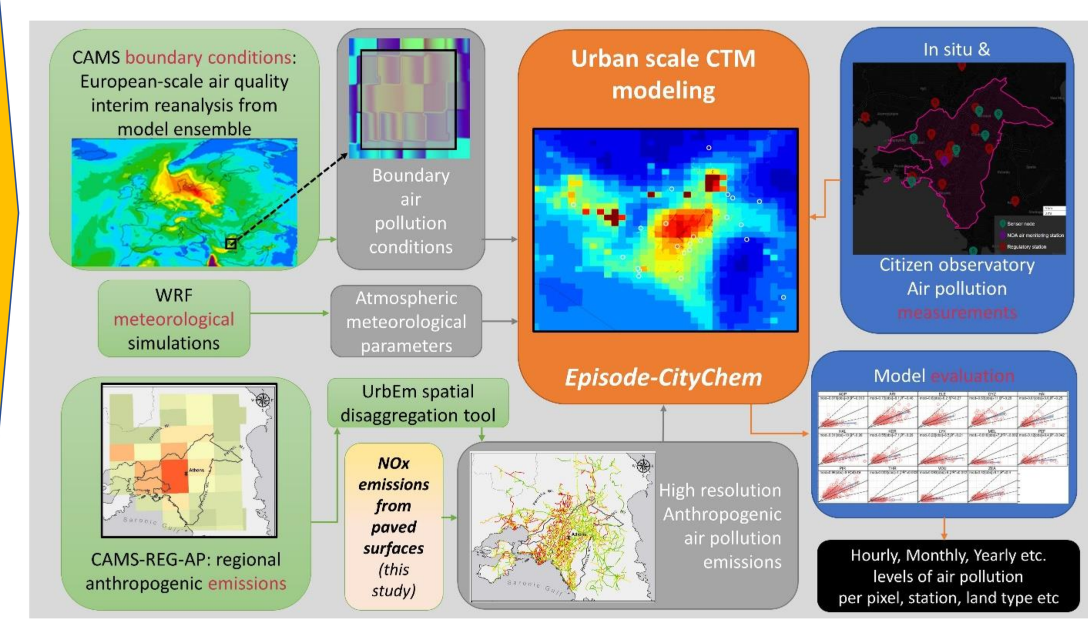


✓ Determination of Emission factors (EFs) versus Temperature, relative humidity, light irradiation.

✓ $EF = C \times 12.187 \times \left(\frac{M}{273.15+T}\right) \times \frac{Q_e}{S_{asphalt}}$

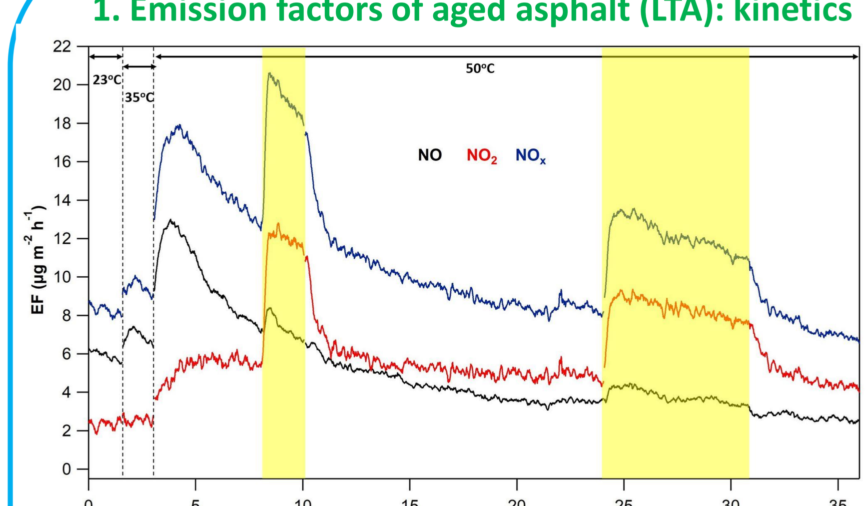
High-resolution numerical atmospheric modelling

✓ Case study: City of Athens, Greece
✓ Considered period: September 2019 (before Covid-19)



Results section – Part 1: NOx emissions

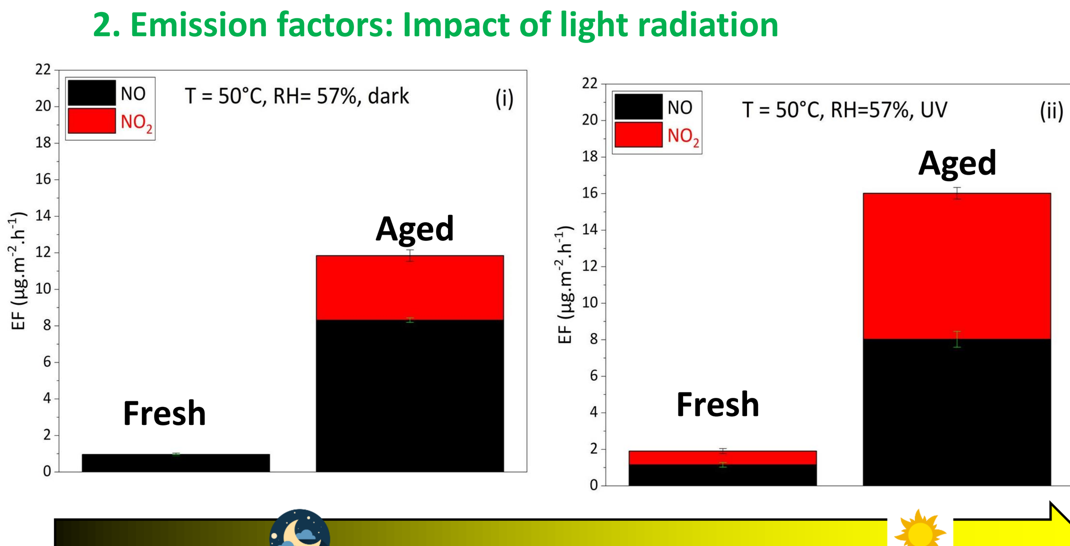
1. Emission factors of aged asphalt (LTA): kinetics



General conclusions (not necessary presented in this graph)

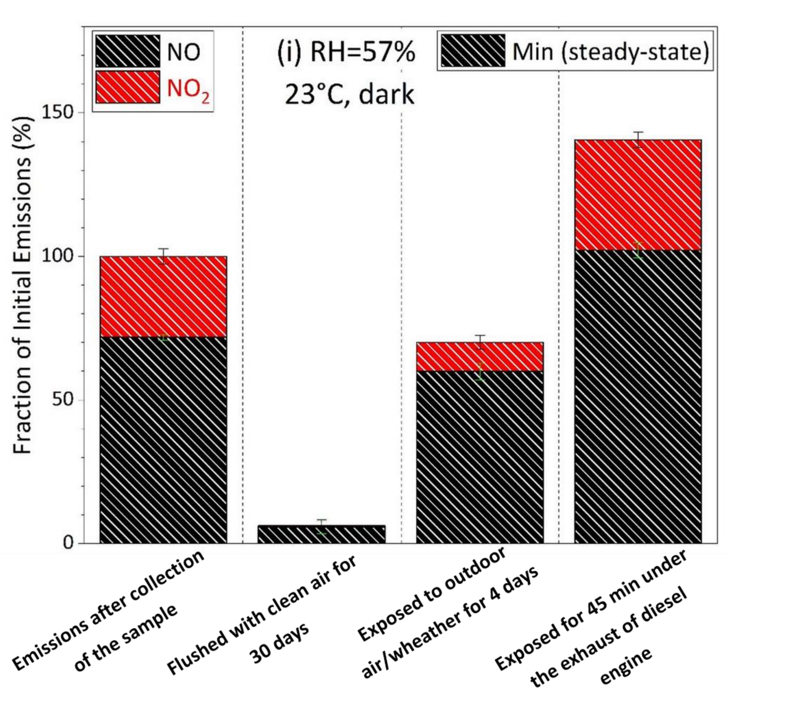
- NOx emissions are stable with time at temperatures of 23°C and 35°C
- At 50°C, NO_2 has steady emissions at each step (plateau), whereas NO generally peaks and then decreases

2. Emission factors: Impact of light radiation

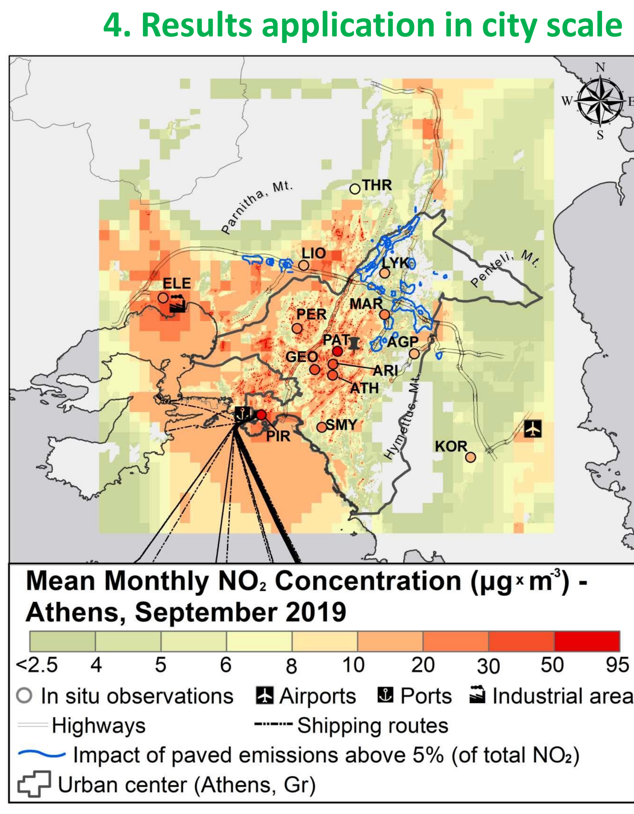


- Fresh asphalt emissions are significantly lower than aged => Asphalt interacts with air pollution. How? NOx, adsorption? Chemical transformations releasing NOx?
- **Light irradiation enhance the formation of NO_2.** Photolysis of organic or inorganic surface species leading to the formation of NO_2?

3. Asphalt surface regeneration experiments



4. Results application in city scale model



In the case of Athens, and the city centre, the contribution of asphalt pavements to total NOx is below 1%.

Outside the city centre, asphalt contribution could be up to 20% of total NOx concentrations.

NOx emissions should be viewed case by case.

Climate change and pollution reduction trends in urban areas, could make the contribution of asphalt pavements more important.

Asphalt pavements do not seem to explain the deficiency of models to predict NOx emissions.

What about NOx deposition?

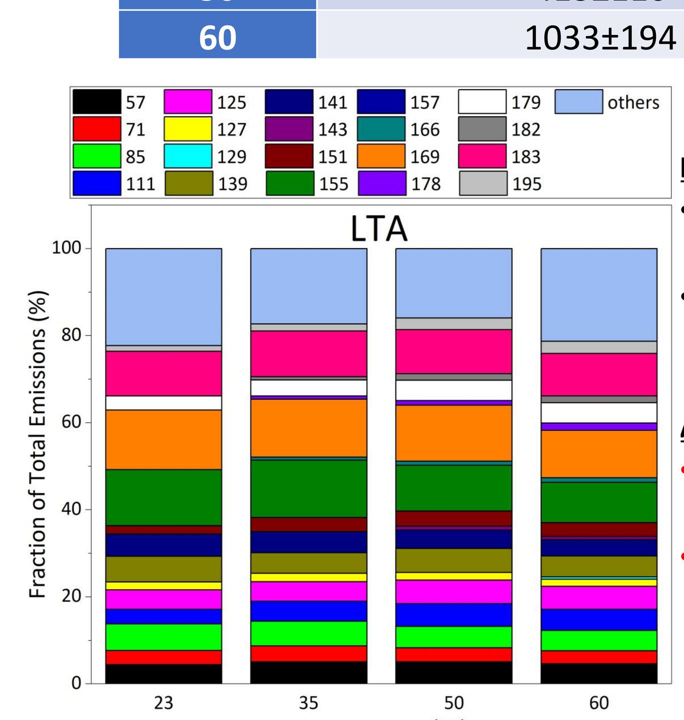
- Asphalt surfaces should be considered as a continuous source of air pollution
- Construction of hourly profiles & implementation in model

Results section – Part 2: VOCs emissions

1. VOCs emissions with PTR-MS (real time monitoring of volatile fraction)

Table 1: EFs determined with PTR-MS under dry and dark conditions

T (°C)	Total VOC EF_{PTR} STA (µg m^{-2} h^{-1})	Total VOC EF_{PTR} LTA (µg m^{-2} h^{-1})
23	2±2	223±49
35	64±29	314±66
50	413±110	466±81
60	1033±194	989±134



Information presented in the graph

- More than 70 masses were identified and quantified. A list of 20 masses contributes to more than 80% of total EFs.
- Under dry and dark conditions, the temperature increases Efs of the same compounds.

Additional information not presented in the graph

- At temperatures below 35°C fresh samples emit less than aged samples. Above that threshold, fresh samples are more emissive.
- VOCs emissions increase with RH and UV light (by a factor of 10)

2. VOCs emissions with TD-GC-FID-MS (1 hour sampling in a Carbotrap 202 tube to monitor off-line the semivolatile, S-VOCs fraction, C_5 to C_{20} species)

Table 2: EFs determined with TD-GC-FID-MS under dry and dark conditions

T (°C)	Total VOC EF_{GC} STA (µg m^{-2} h^{-1})	Total VOC EF_{GC} LTA (µg m^{-2} h^{-1})
23	3±1	53±18
35	14±4	51±18
50	109±34	129±44
60	222±70	186±63

3. Atmospheric implications

➢ **In terms of total VOCs emissions**

Under dry and dark, VOCs emissions are not important compared to total VOCs in urban areas. However, under humid and light irradiation they can be an important source

➢ **In terms of Ozone and Particles formation efficiency**

Under evaluation: the release of high levels of unsaturated compounds could make asphalt an important source of secondary pollutants.
The implementation of high resolution city scale atmospheric model experiments is necessary for a thorough evaluation

Perspectives

- For NOx emissions from asphalt pavements: Consider future scenarios and evaluate their contribution in future air quality. A good approach could be the COVID-19 restricted periods
- For VOCs emissions from asphalt pavements: Need for a detailed and speciated analysis to identify and quantify higher number of VOCs. Need to evaluate their current and future contribution to urban air quality (i.e. contribution to total VOCs and VCPs, impact on Particles and ozone formation). Both experimental and modelling studies are necessary.

Acknowledgements

This work was supported by the French national program LEFE/INSU within the framework of the EMAPI project and the IMT Nord Europe in the framework of IRAPAQ projects. The atmospheric numerical work is largely performed in the frame of the EU H2020 project e-shape (grant agreement 820852).