Estimation of city-scale NO_x emission flux using the GEMS dataset

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Background and Motivation

1) The need for estimating NO_x emissions in a sudden event (e.g., COVID-19)

Anthropogenic emissions of NO_x are primarily generated from combustion activities by power plants and vehicles. Once a sudden event occurs, it is essential to quickly identify and provide information on changes in emission levels in major NO_x-emitting cities with dense populations.

2) Slow development of bottom-up inventories

Estimating emissions by assessing the total fuel consumption from each NO_x-emitting source is time-consuming. If information on fuel consumption is not available, emission data cannot be obtained. Emissions from unknown anthropogenic sources cannot be identified.

3) The existence of geostationary satellite measurement

Due to the insufficient data from polar-orbiting satellites, it is challenging to estimate annual/seasonal emissions. But GEMS observes the East Asian region (5000×5000 km^2) ~6 to 10 times a day. Therefore, when estimating NO_x emissions in the East Asian region, the usage of GEMS data is more beneficial as it reduces the uncertainty due to sampling.

Previous study: Estimation of NO_x emission using LEO satellites

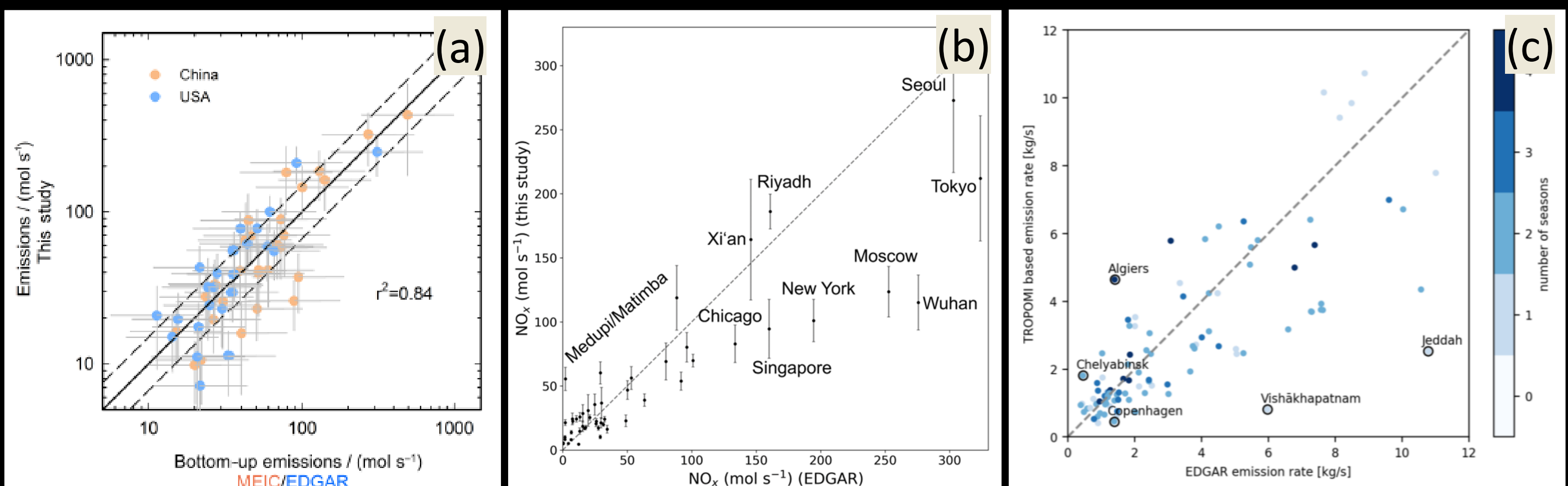
Several previous studies (e.g., Beirle et al., 2011; Liu et al., 2016) succeeded to estimate city-scale NO_x emissions using OMI or TROPOMI satellite data, by analyzing the differences in NO_2 distribution between calm and windy condition.

(a) Emission estimates for 53 cities in China and the United States using OMI data in the ozone season during 2005-2013. (Liu et al., ACP, 2016)

(b) Emission estimates for 100 cities worldwide using TROPOMI data during 2018-2020. (Lange et al., ACP, 2022)

(c) Emission estimates for 100 cities worldwide using TROPOMI data during 2018-2021. (Beirle et al., EGUsphere, 2024)

(x-axis: bottom-up, y-axis: satellite-based estimation)



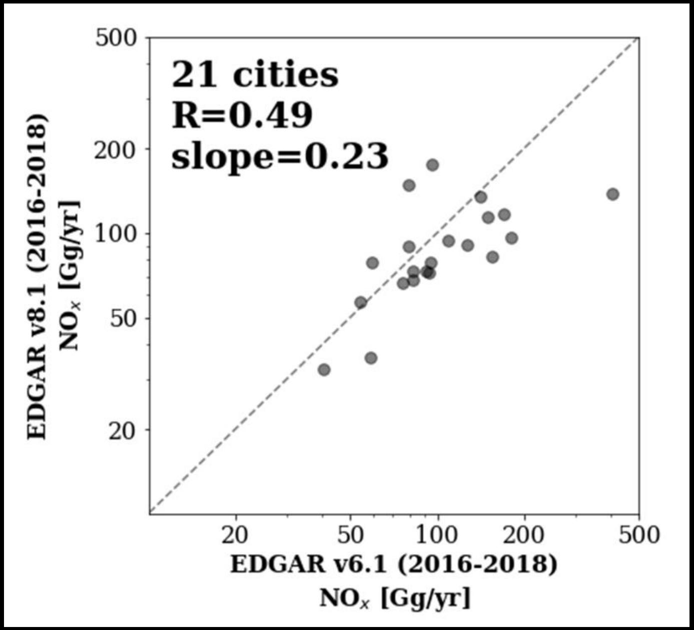
Data description

	Satellite	Reanalysis	Bottom-up inventory	
	GEMS L2 v2.0	ECMWF ERA5	EDGAR v6.1	EDGAR v8.1
Period	03.01.2022 – 02.28.2023	03.01.2022 – 02.28.2023	2016-2018	2016-2022
Variable	tropospheric NO_2 VCD (cloud fraction < 0.5, SZA < 70°)	u, v component (averaged at 1000hPa and 975hPa)	Total NO_x emissions	
Resolution	hourly, 7×8 km^2	hourly, 0.25°×0.25°	Annual, 0.1°×0.1°	

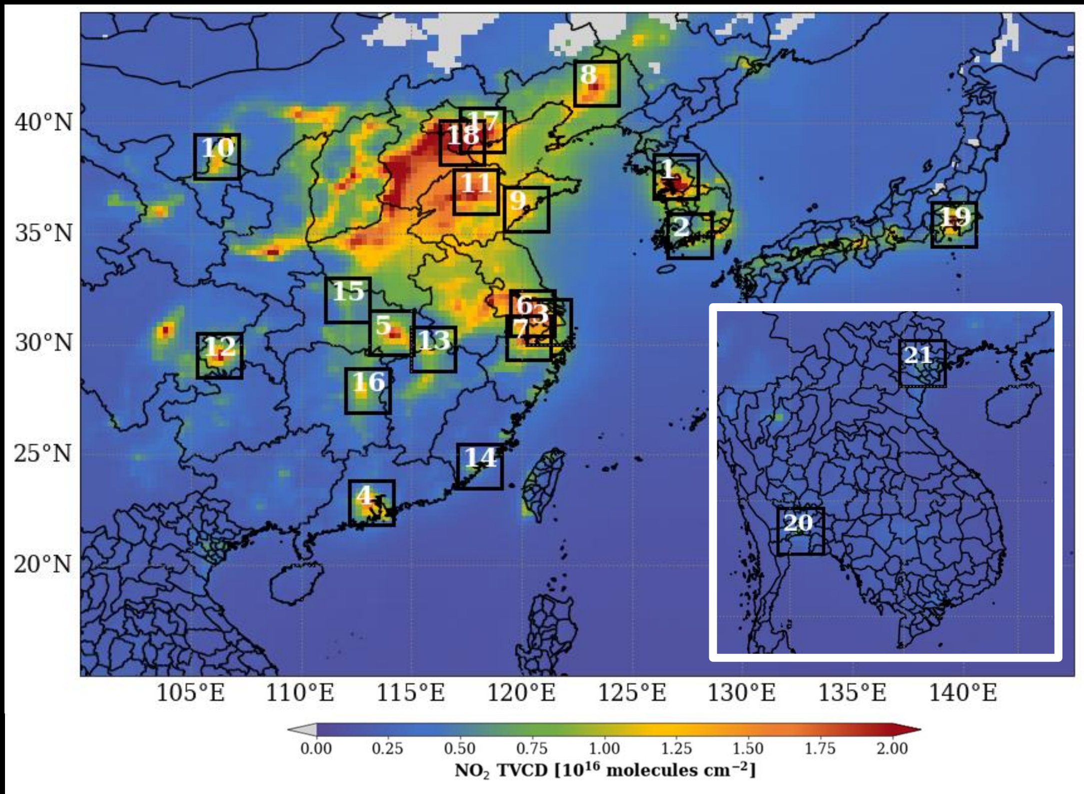
Collocation : To classify satellite data based on wind direction and wind speed

- Spatial collocation : GEMS data was regridded to a resolution of 0.05°×0.05°, resulting in 25 GEMS data grid cells being included within a single grid cell of the ERA5 data. 25 GEMS data grid cells are averaged for the same day under specific wind direction and wind speed conditions.
- Temporal collocation : we matched the time of GEMS observation with that of ERA5. (e.g. GEMS 03:50 UTC ⇔ ERA5 04 UTC)

EDGAR (v6.1 versus v8.1)



Target area



City		
1. Seoul	2. Yeosu	South Korea
3. Shanghai	4. Guangzhou	China
5. Wuhan	6. Suzhou	China
7. Hangzhou	8. Shenyang	China
9. Qingdao	10. Yinchuan	China
11. Jinan	12. Chongqing	China
13. Jiujiang	14. Xiamen	China
15. Xiangyang	16. Changsha	China
17. Tangshan	18. Tianjin	China
19. Tokyo		Japan
20. Bangkok		Thailand
21. Hanoi		Vietnam

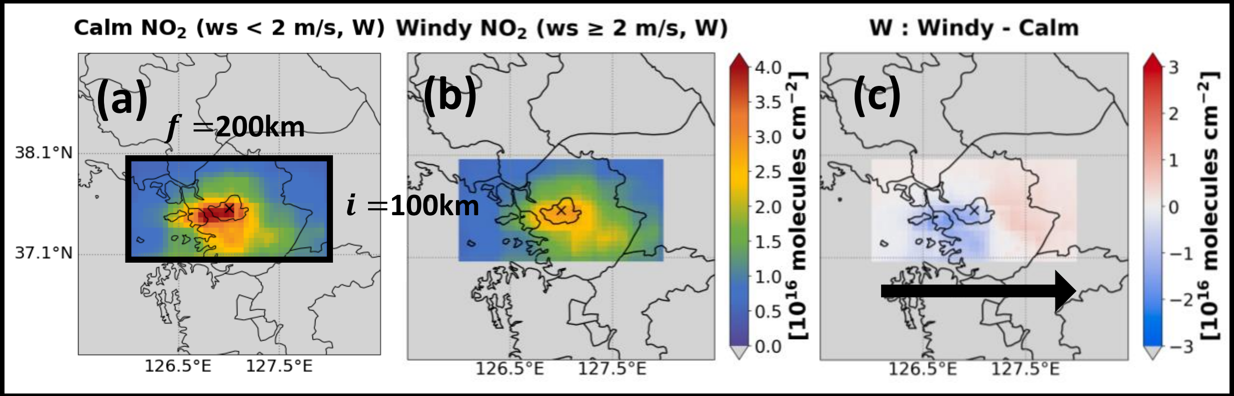
- 21 cities are selected which have high NO_2 and located in GEMS scan area
- South Korea and Japan : Cities with high NO_2 concentrations are selected.
- China : Cities are selected as study area in previous studies. (Liu et al., 2016; Xue et al., 2022)
- Thailand and Vietnam : The capitals of each country are selected because of significant NO_x emissions and minimal elevation difference.

Methodology

(Main reference: Liu et al., 2016)

1) Mean tropospheric NO_2 VCD (vertical column density) distribution

- All NO_2 data within the analysis period are categorized by wind direction and wind speed for each grid, and then averaged (Cloud screening: cloud fraction < 30%).



wd	N, S, E, W, NE, SW, NW, SE
ws	Calm wind / Windy condition (threshold : 2 m/s)

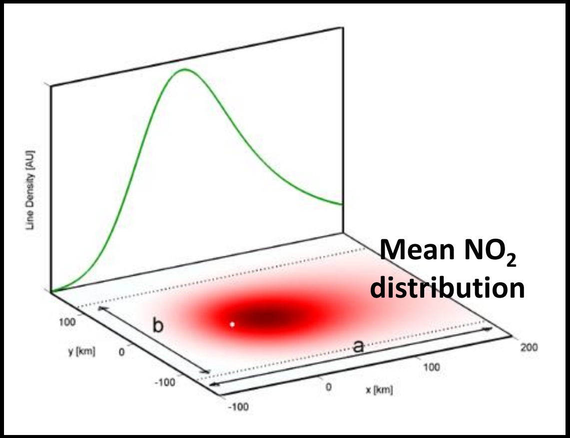
(a) : calm wind condition

(b) : windy condition

(c) : (b)-(a)

2) NO_2 line density

- A curve calculated by integrating the mean NO_2 distribution along a direction (b) perpendicular to a specific wind direction (a).
- NO_2 line density is obtained under each wind direction and wind speed condition.
- A peak appears at the location where the emission source is located.

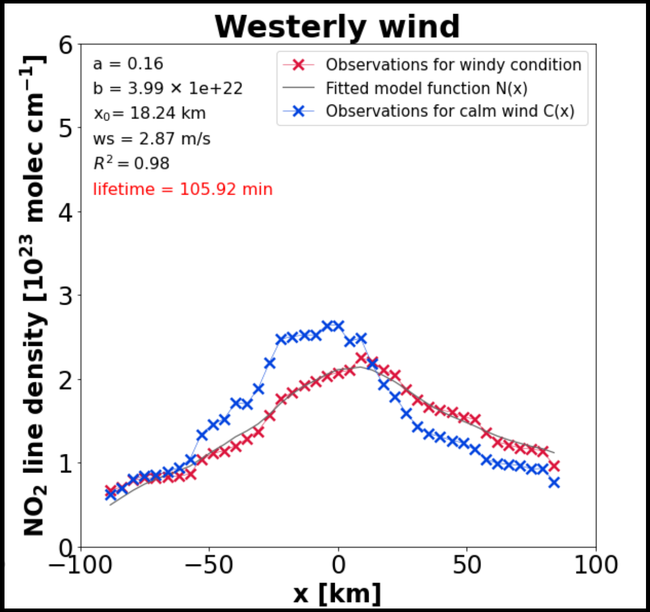


Beirle et al. (2011)

Methodology

3) Effective NO_x lifetime

- NO_2 line density calculated under calm wind conditions and windy conditions is used.
- A nonlinear least squares fit is performed by substituting the NO_2 line density under calm wind conditions for C(x) and parameters (a, b, x_0) in the model function N(x).
- When the fitted N(x) takes the shape of the line density under windy conditions, a, b, and x_0 are fitted and provide information about the NO_2 distribution that changed by the wind.
- Specifically, x_0 represents the distance the plume has been transported (e-folding distance), and by substituting it into eq. (3), the effective lifetime (τ) can be calculated.



$$\mathrm{N}(x) = a \times (e \otimes C)(x) + b \qquad \text{---------------------- (1)}$$

$$e(x) = \exp\left(-\frac{x-X}{x_0}\right) \text{ for } x \geq X,\ 0\ otherwise \qquad \text{--- (2)}$$

$$\tau = \frac{x_0}{w}, \qquad w = w_{windy} - w_{calm} \qquad \text{------------- (3)}$$

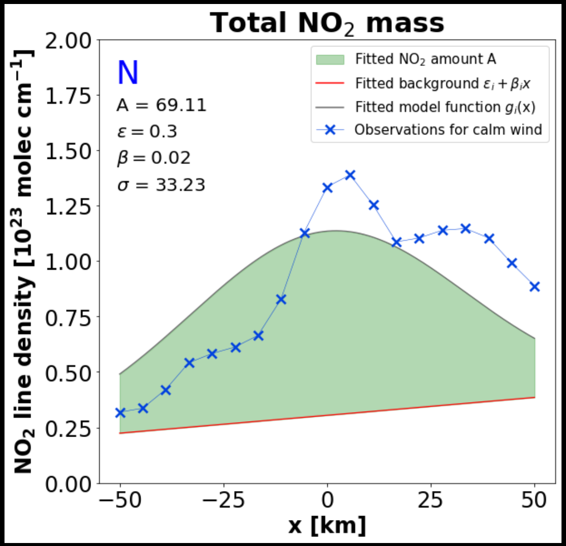
a : scaling factor b : offset x_0 : e-folding distance w : mean wind speed

⇒ Transport, chemical decay, and spatial smoothing are not considered separately. So τ is not exactly same with lifetime in the real atmosphere.

Methodology

4) NO_x total mass

- To minimize interference caused by the advection, only the NO_2 line density under calm wind conditions is used.
- The NO_2 line density is calculated for a 40×100 km^2 area in 21 cities, and line densities under 8 wind directions are fitted by the EMG (Exponentially Modified Gaussian) function from eq. (4).
- Fitted A means NO_2 total mass excepting background NO_2 ($\varepsilon_i + \beta_i x$). (detailed in next page)
- Scaling NO_2 to NO_x : To calculate NO_x total mass, fitted A is multiplied by $[NO_x]/[NO_2]$ ratio (=1.32).
- **NO_x emission rate (NO_x amount per unit time) = NO_x total mass / lifetime (τ)**

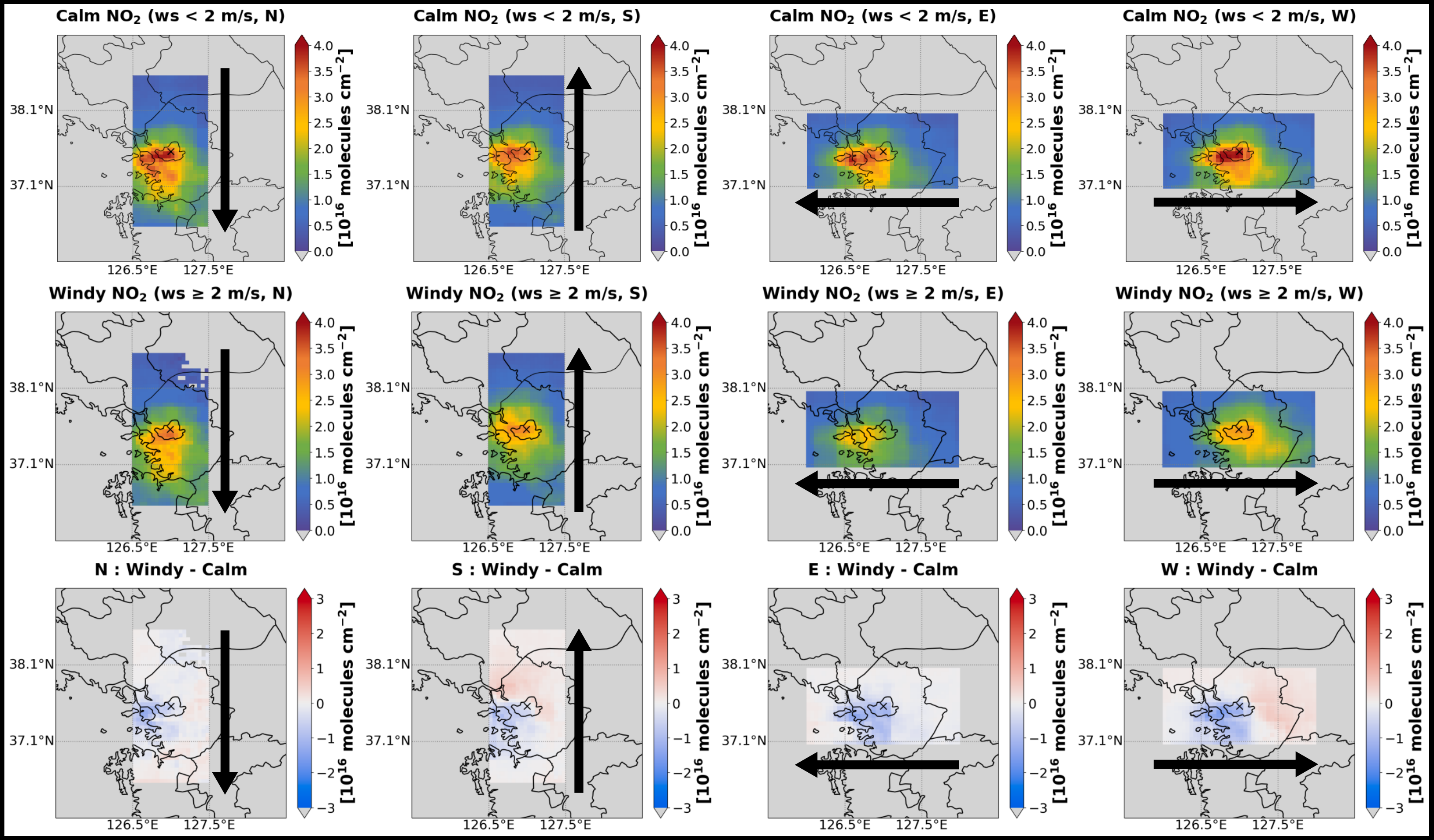


$$g_i(x) = A \times \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{(x-X)^2}{2{\sigma_i}^2}\right) + \varepsilon_i + \beta_i x \qquad \text{----- (4)}$$

Blue line : NO_2 line density in the calm wind condition
Red line : background NO_2 ($\varepsilon_i + \beta_i x$) - Result of linear regression using NO_2 line density calculated up to the 5th percentile
Gray line : fitted $g_i(x)$
Green area : fitted A = NO_2 total mass

Results : Mean NO_2 distribution

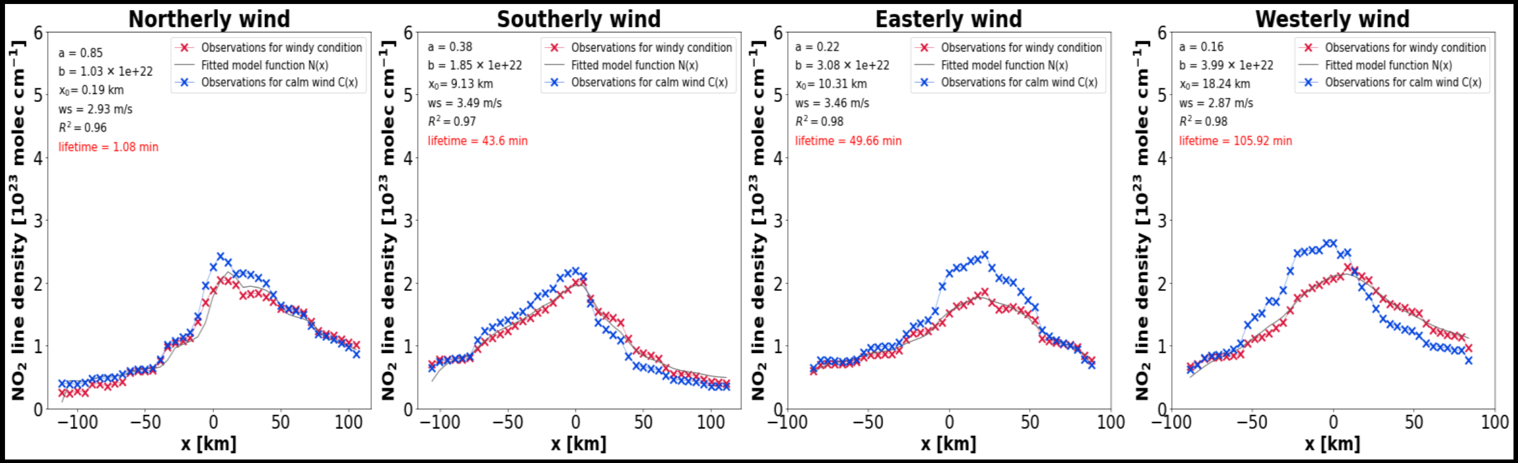
(a)
(b)
(c)



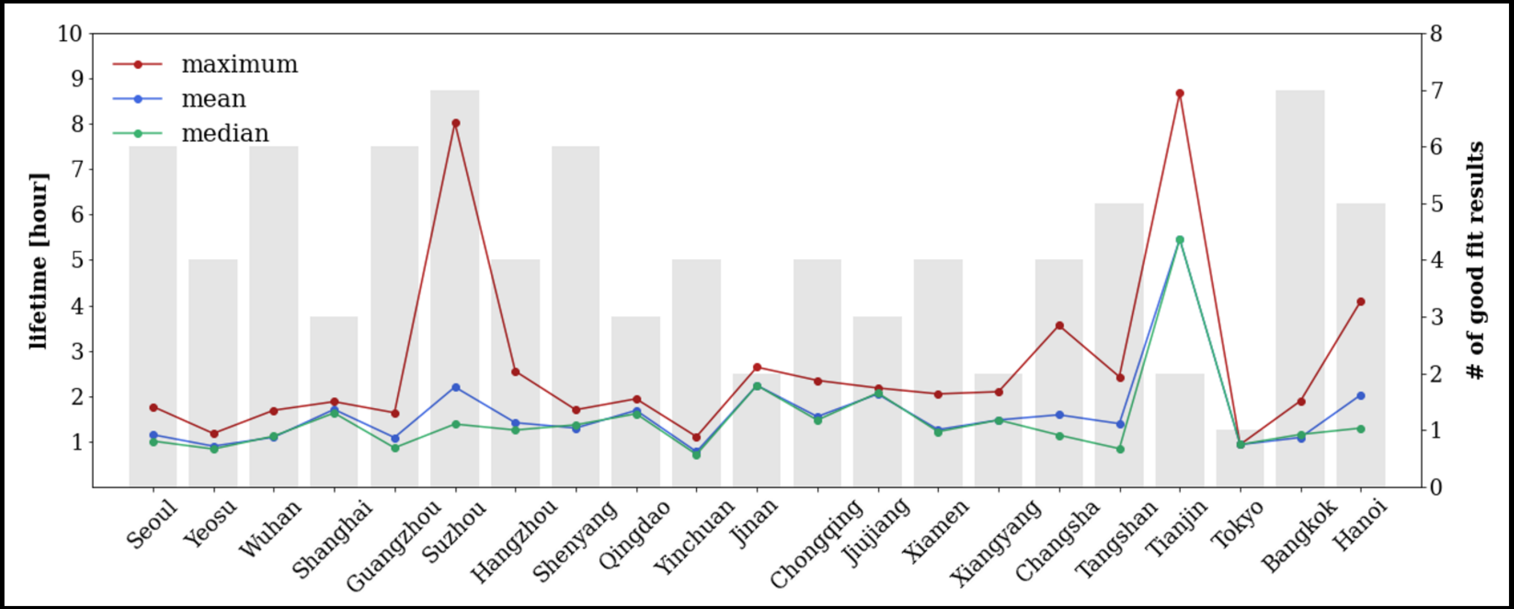
- (a) : The result under calm wind condition for N, S, E, W wind.
- (b) : The result under windy condition for N, S, E, W wind.
- (c) : (b)-(a). According to (c), the movement of the NO_2 plume in the troposphere can be observed following the wind direction (black arrows).
- (a) and (b) are used to calculate line densities.

Results : effective NO_x lifetime

Fitting result under N, S, E, W wind in Seoul, 2022

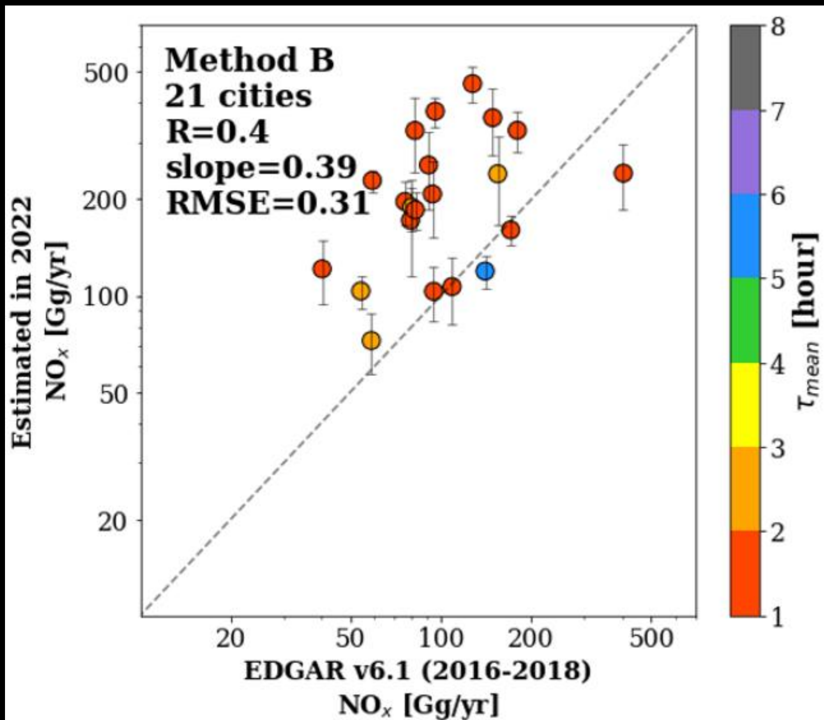


Estimated effective e-folding lifetime (Maximum, mean, and median among total 8 windy cases)

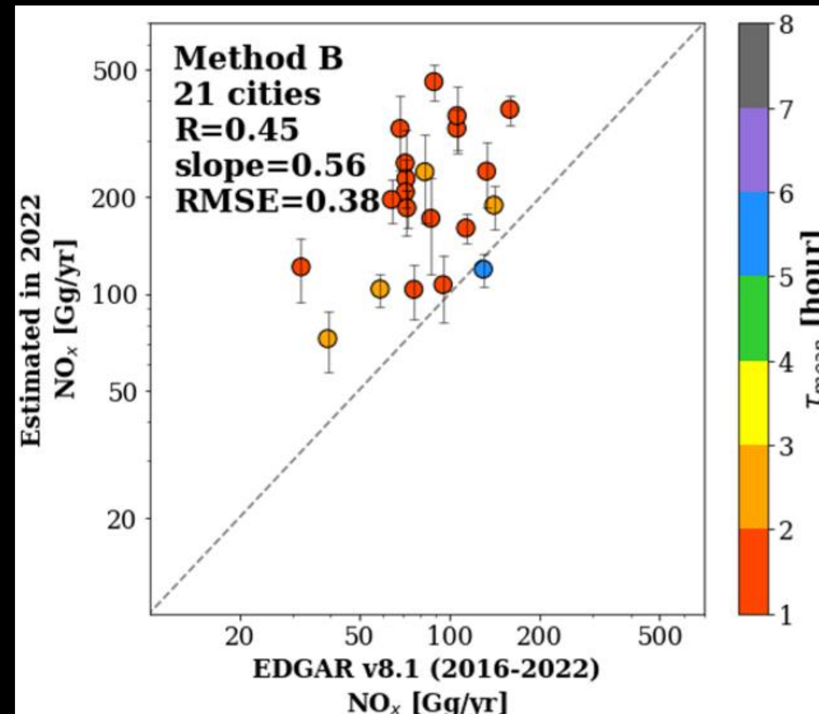


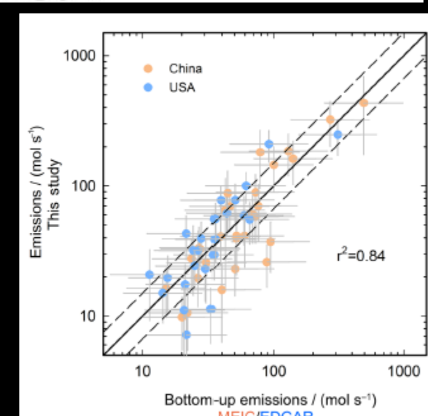
Results : NO_x emission rate (compared with EDGAR)

With EDGAR v6.1 (mean 2016-2018)



With EDGAR v8.1 (mean 2016-2022)





- Positive correlation is found between the EDGAR and our GEMS-based estimation.
- GEMS-based NOx emission shows the overestimation. This will be redeemed better when new GEMS NO_2 product is applied.

Concluding remarks with my private curiosity

Reducing the GEMS (and TEMPO) NO_2 vs. Enhancing the TROPOMI NO_2

- I am not able to say which one is better, which one is not. (Qualitatively all products are good to use for the air quality diagnosis. But quantitatively?)

- In the context of methodology used in this study,
  - GEMS NO_2 VCD - based estimated NO_2 emission is larger than EDGAR NO_2 emission (my work)
  - TROPOMI NO_2 VCD - based estimated NO_2 emission is comparable to EDGAR NO_2 emission (e.g., Liu et al., 2016)

- And new version (improvement assumed) of EDGAR (ver 8.1) shows lower emission than old version (ver 6.1) (At least in the range of this study. Need to see more).

Thanks for your attention