

Atmospheric observations of CO₂, ¹⁴CO₂ and O₂ concentrations to capture fossil fuel CO₂ emissions from the Greater Tokyo Area

Yukio Terao¹ (¹⁴CO₂ and CO₂/CH₄/CO at TST, YYG, NIES and NDA)

Yasunori Tohjima¹ (O₂ and CO₂ at TST and NIES)

Shigeyuki Ishidoya² (O₂ and CO₂ at YYG)

Mai Ouchi¹, Yumi Osonoi¹, Hitoshi Mukai¹, Toshinobu Machida¹ (¹⁴C, NIES lab., TST)

Hirofumi Sugawara³ (CO₂ flux at YYG, CO₂/CH₄/CO at NDA)

Naoki Kaneyasu² (aerosol component at YYG)

Yosuke Niwa¹ (NICAM atmospheric transport model)

¹ National Institute for Environmental Studies (NIES), Japan (yterao@nies.go.jp)

² National Institute of Advanced Industrial Science and Technology (AIST), Japan

³ Department of Earth and Ocean Science, National Defense Academy (NDA), Japan

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Points:

- ✓ We have performed ground-based atmospheric observations for measuring concentrations of CO_2 , $^{14}\text{CO}_2$ and O_2 in the Greater Tokyo Area.
- ✓ The $^{14}\text{CO}_2$ measurement was used for separating the fossil fuel CO_2 emissions from the biotic emissions. The $\text{O}_2:\text{CO}_2$ exchange ratio (oxidation ratio, OR) was used for the partitioning of CO_2 into emissions from gas fuels and gasoline.
- ✓ Results from $^{14}\text{CO}_2$ measurements showed that a ratio of fossil fuel-derived CO_2 to the variation of CO_2 concentrations was 71% (TST) and 73% (YYG) in average for winter but varied from 44% to 92%, indicating significant contribution of biotic CO_2 in Tokyo.
- ✓ Results from OR showed larger OR in winter than in summer (due to both wintertime increases of fossil fuel combustion and summertime terrestrial biospheric activities) at TST and YYG and larger OR in the morning and late evening in winter due to increase of gas fuel combustion at YYG.
- ✓ Our colleague developed building/road-scale dynamic CO_2 mapping and grid-based high spatial resolution CO_2 emission inventory in Tokyo.

$^{14}\text{CO}_2$ and O_2 observations to capture fossil fuel CO_2 emissions

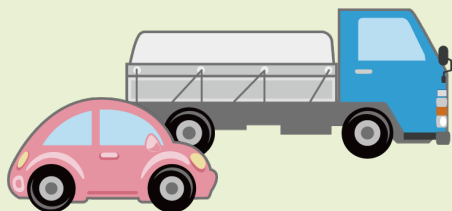
CO_2 from natural gas

$^{14}\text{CO}_2 = \text{zero}$, $\text{OR}^* = 1.97$



CO_2 from gasoline

$^{14}\text{CO}_2 = \text{zero}$, $\text{OR} = 1.5$



*Oxidative Ratio; $-\text{O}_2/\text{CO}_2$ (mol/mol)

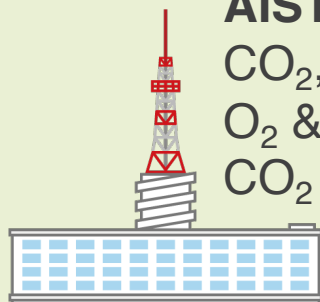
Yoyogi site (52m)

AIST, NDA, NIES

CO_2 , CH_4 , CO , ...

O_2 & $^{14}\text{CO}_2$

CO_2 & heat flux



Biospheric CO_2

$^{14}\text{CO}_2$ included, $\text{OR} = 1.1$
(plants) & 1.25 (human)



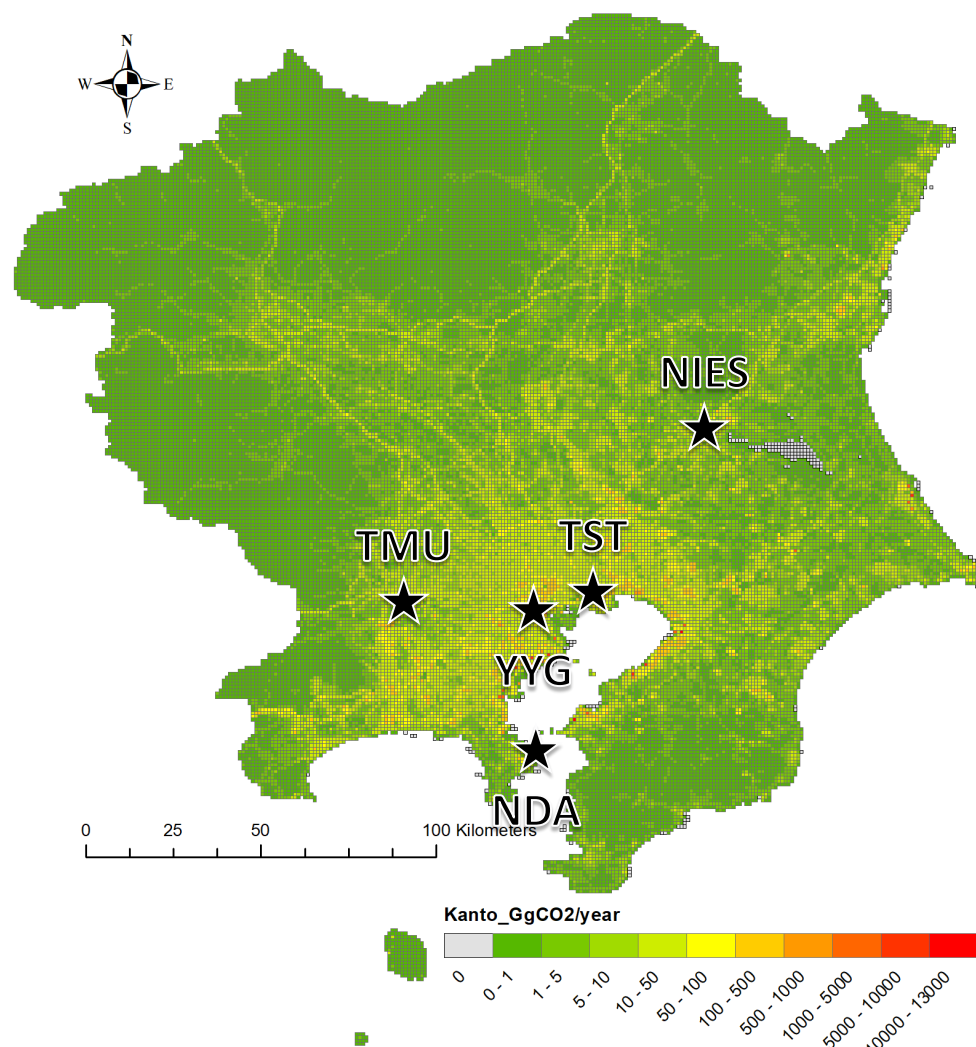
NIES Tokyo Skytree observatory (250m)

CO_2 , CH_4 , CO , ...

O_2 & $^{14}\text{CO}_2$



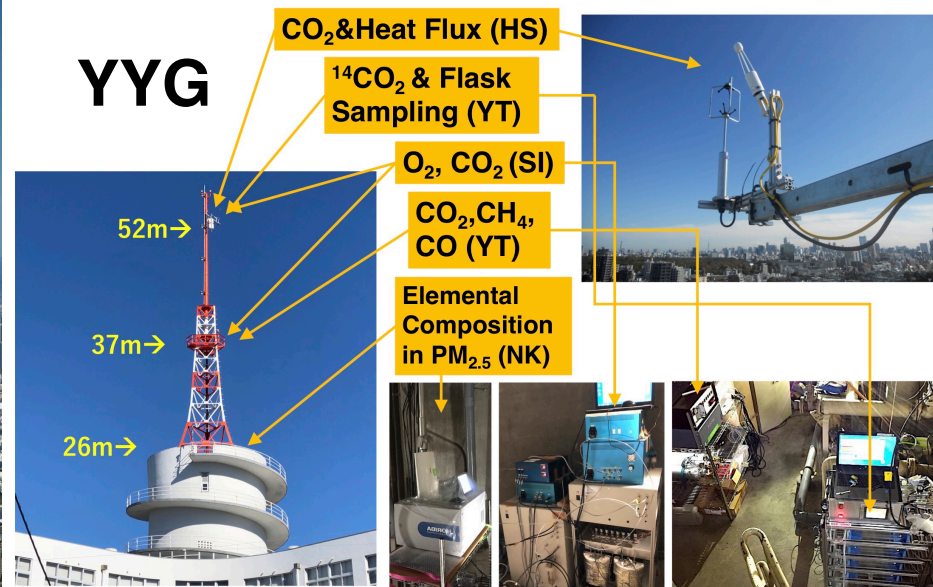
Atmospheric Observations in the Greater Tokyo Area



Tokyo SkyTree (TST)	Sumida, Tokyo	Continuous CO ₂ , CH ₄ , CO and O ₂ Flask (¹⁴ CO ₂)
Yoyogi (YYG)	Shibuya, Tokyo	Continuous CO ₂ , CH ₄ , CO and O ₂ Flask (¹⁴ CO ₂) CO ₂ flux, aerosol
NDA	Yokosuka, Kanagawa	Continuous CO ₂ , CH ₄ , CO
NIES	Tsukuba, Ibaraki	Continuous CO ₂ , CH ₄ , CO and O ₂ Flask (¹⁴ CO ₂)
Tokyo Metropolitan Univ. (TMU)	Hachioji, Tokyo	Continuous CO ₂

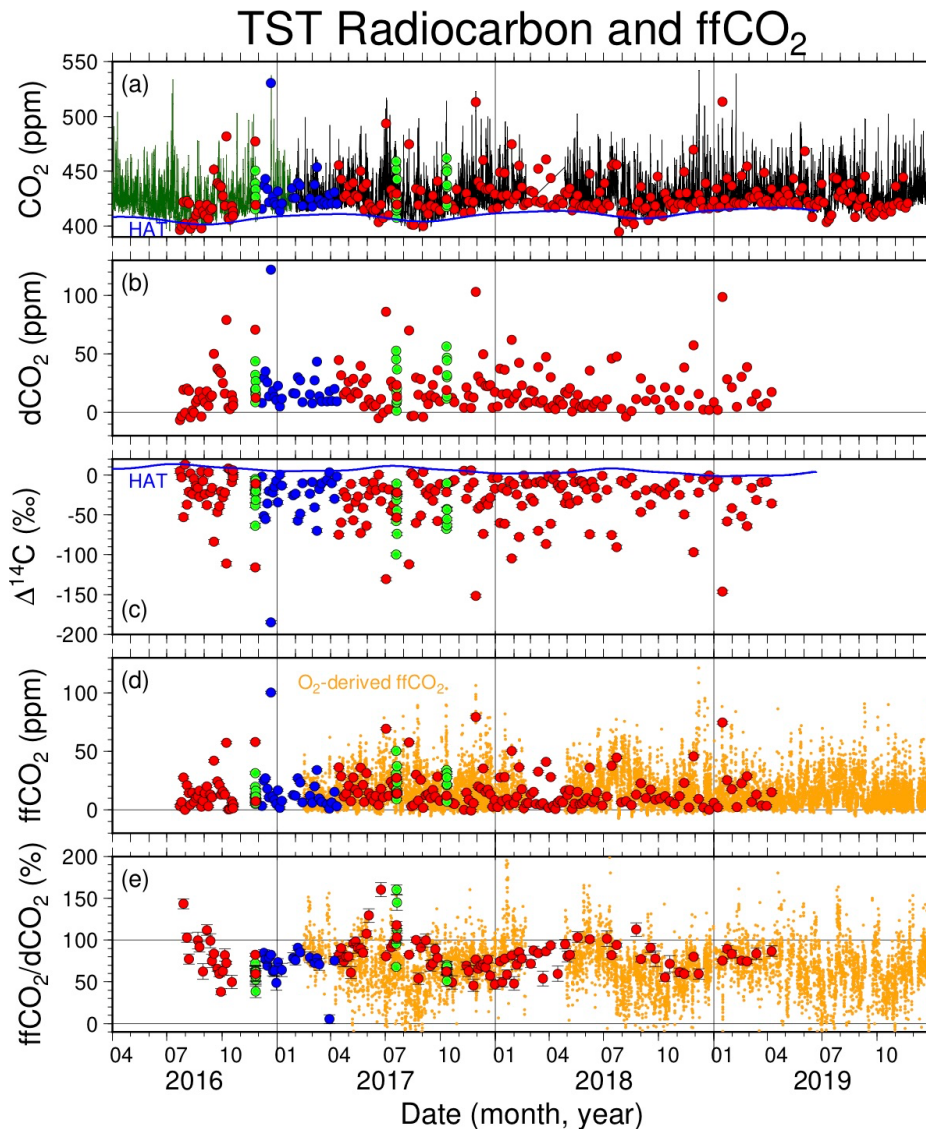
Location of atmospheric observation sites in the Greater Tokyo Area. Color map shows a 1km mesh CO₂ emission data in 2005 (updated from Cong et al., CBM, in review).

Atmospheric Observations at TST and YYG



	Tokyo SkyTree (TST)	Tokai University, Yoyogi (YYG)
$\text{CO}_2, \text{CH}_4, \text{CO}$	CRDS (Picarro G2401)	
$\text{O}_2 \& \text{CO}_2$	Fuel cell O_2 analyzer (Oxzilla II) and NDIR (LI-840A) (Hoshina et al., 2018)	Paramagnetic O_2 analyzer (POM-6E) and NDIR (LI-820) (Ishidoya et al., 2017)
$^{14}\text{CO}_2$	Flask sampling of whole air (every 4-day). ^{14}C analysis by NIES-CAMS.	
CO_2 flux	N/A	Sonic anemometer (WindMasterPro) and open-path NDIR (LI-7500) (Hirano et al., 2015)
Aerosol	N/A	XRF analyzer (PX-375) (Kaneyasu et al., 2020)

Fossil fuel CO₂ estimated from ¹⁴CO₂ and O₂ at TST



(a) CO₂ mole fraction

Continuous (line) and flask (circles)
measurements (14 p.m. or 20 p.m. LT)
Background (Hateruma Island) (curve)

(b) CO₂ increase from background (ΔCO_2)

(c) $\Delta^{14}\text{C}$ in CO₂

$$\Delta^{14}\text{C} = \delta^{14}\text{C} - 2(\delta^{13}\text{C} + 25)(1 + \delta^{14}\text{C}/1000)$$

(d) CO₂ from fossil fuel combustion (CO₂^{ff})

derived from $\Delta^{14}\text{C}$ (red/blue circles)

$$\text{CO}_2^{\text{ff}} = \text{CO}_2^{\text{obs}} (\Delta^{14}\text{C}^{\text{bg}} - \Delta^{14}\text{C}^{\text{obs}}) / (\Delta^{14}\text{C}^{\text{bg}} + 1000)$$

and from O₂ (orange dots)

$$\text{CO}_2^{\text{ff}} = (\Delta\text{O}_2 + \alpha_{\text{bio}} \cdot \Delta\text{CO}_2) / (\alpha_{\text{bio}} - \alpha_{\text{ff}})$$

ΔO_2 and ΔCO_2 : change from baseline

α_{bio} : Oxidative ratio for biosphere
(1.05 ± 0.05)

α_{ff} : Oxidative ratio for fossil fuel combustion
from inventory (1.68 ± 0.03)

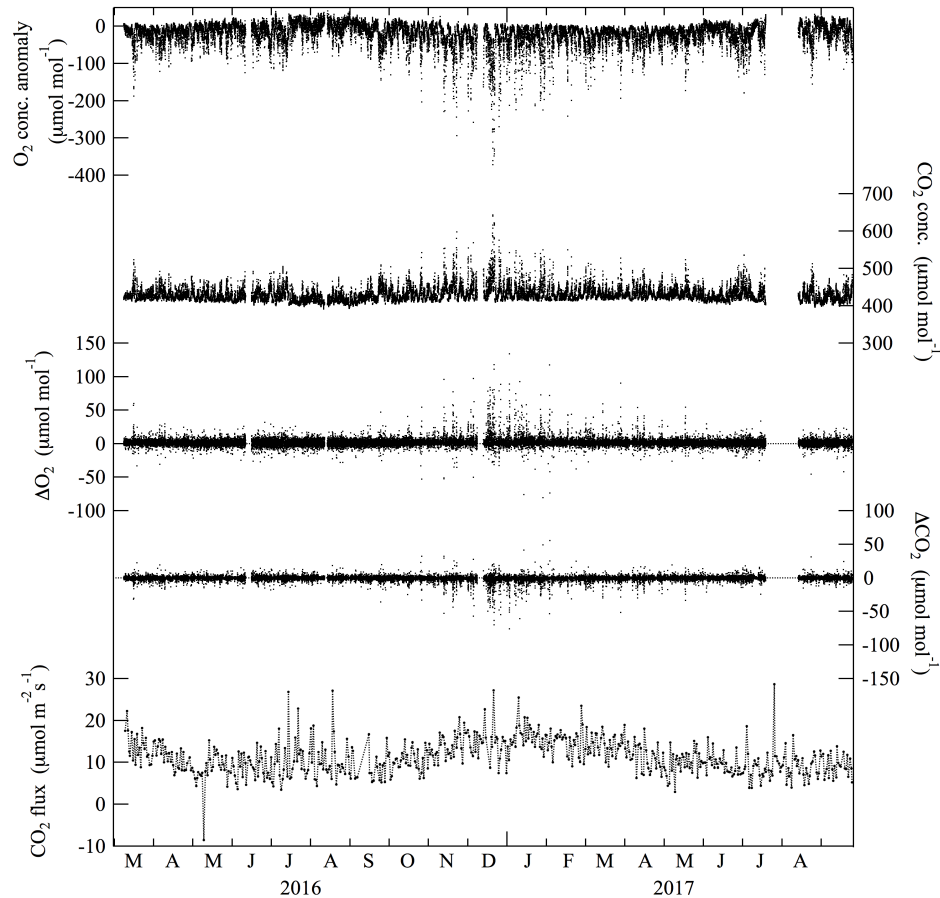
(e) CO₂^{ff}/ΔCO₂

$\Delta^{14}\text{C}$ -derived CO₂^{ff} was 71% in ΔCO₂ for winter, indicating significant contribution of biotic CO₂ in Tokyo. ($\Delta^{14}\text{C}$ method doesn't work well in summer)

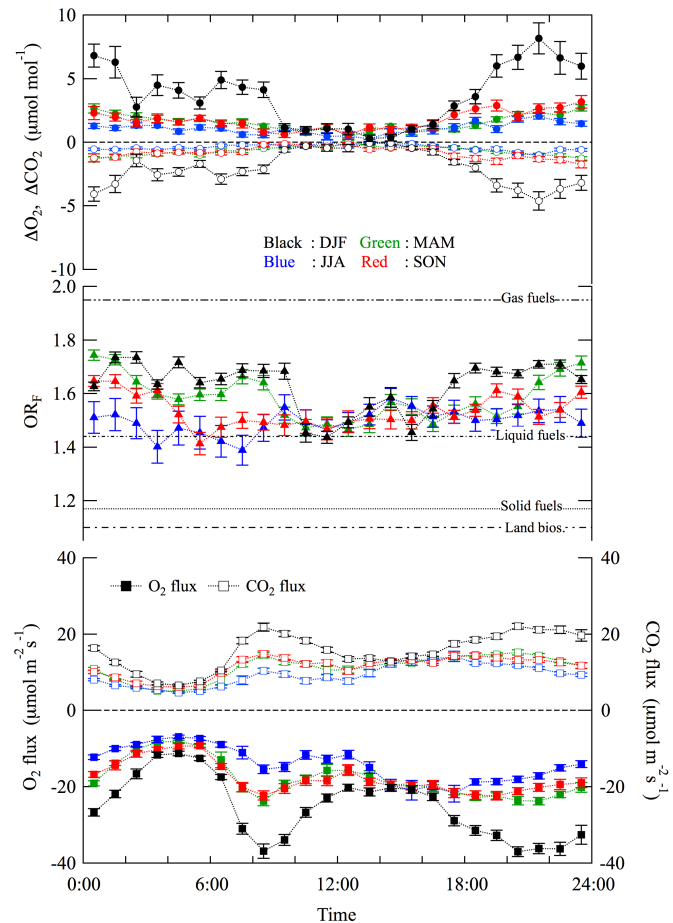
O₂ might be used as a proxy for continuous monitoring of fossil fuel CO₂ content by assuming typical ratio of gas fuels and gasoline combustions.

Oxidative Ratio for net turbulent flux observed at YYG

(Ishidoya et al., ACPD, 2019)



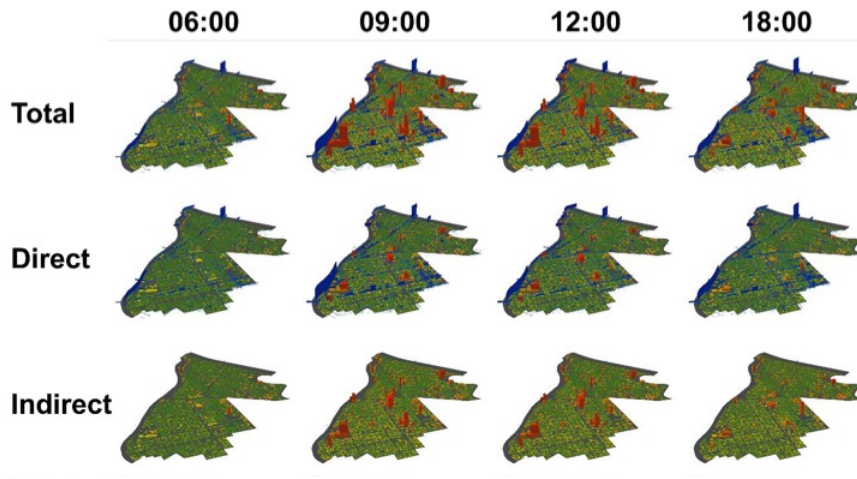
Time series of O_2 and CO_2 concentrations observed at 52 m, ΔO_2 and ΔCO_2 (52 m minus 37 m), and daily mean CO_2 fluxes observed using the eddy correlation method at YYG.



Average diurnal cycles of ΔO_2 and ΔCO_2 for each season: December to February (black), March to May (green), June to August (blue) and September to November (red).

Development of emission inventory in Tokyo

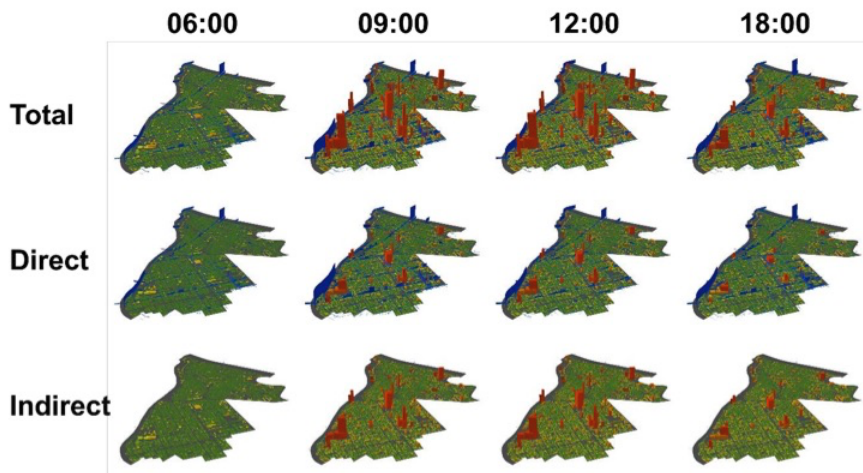
(a) January



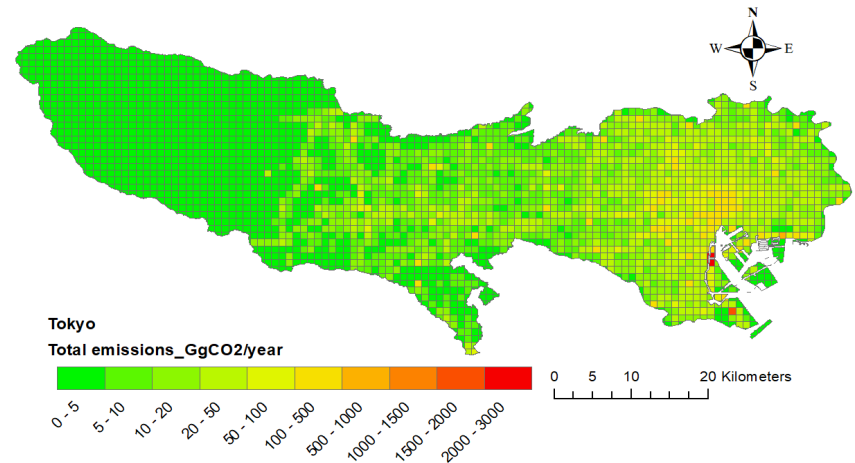
← Building/road-scale dynamic CO₂ mapping developed by Y. Yamagata, T. Yoshida and D. Murakami (NIES)

↓ Grid-based high spatial resolution CO₂ emission inventory developed by R. Cong, M. Saito, A. Ito (NIES) and T. Oda (NASA)

(c) July



Carbon mapping in Sumida, Tokyo
(Yamagata et al., Sustainability, 2018).



CO₂ emission data (1 km mesh) in Tokyo in 2005
(Cong et al., CBM, in review).