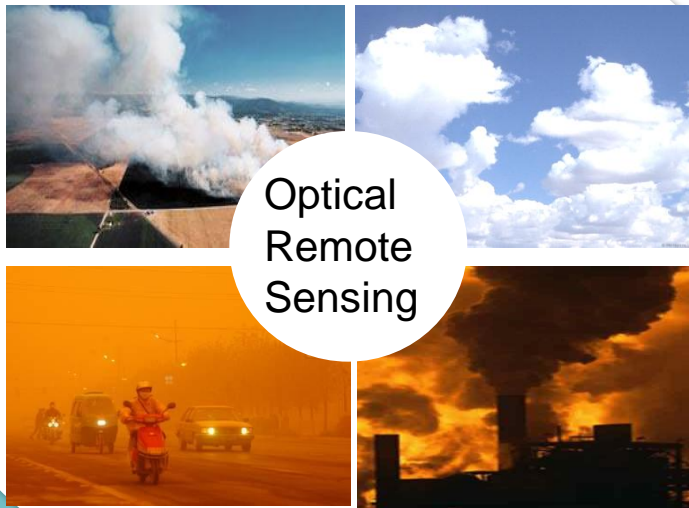


Optical remote sensing of atmospheric trace gases and aerosol



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School of Environmental Science and Engineering
Gwangju Institute of Science and Technology(GIST)*

21 October 2010

Outline

- Introduction
- DOAS principle and methods
- Examples of measurements
 - I. MAX-DOAS measurements of ClO, SO₂, and NO₂
 - II. Imaging DOAS measurements of NO₂
 - III. MAX-DOAS measurement of Aerosol
 - IV. Satellite measurement of NO₂ and SO₂
- Summary



INTRODUCTION

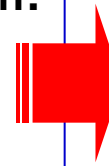


Effects of Atmospheric Aerosols

1. **Human health** : Adverse effects on respiratory and cardiovascular systems
2. **Air quality** : a criteria pollutant, impairment of visibility [Kim and Kim, 2003]
3. **Climate change** : radiative forcing by direct and indirect effects, precipitation

An Integrated Approach for Effective Monitoring of Atmospheric Environment at GIST/ADEMRC

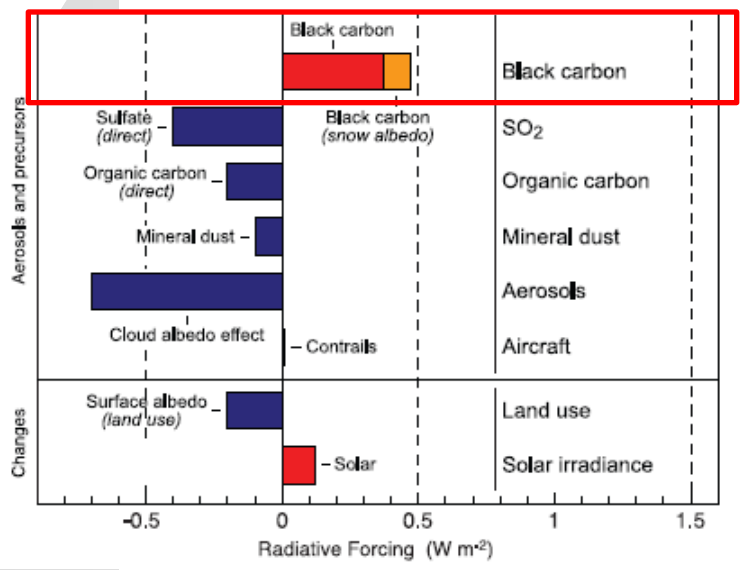
- **Multi-wavelength Raman LIDAR** : aerosol vertical profile, LIDAR ratio, extinction coeff.
- **Sunphotometer** : column aerosol optical depth
aerosol size distribution
- **Satellite retrieval** : spectral aerosol optical depth



**Better
characterization
of physical and
optical properties
of atmospheric
aerosol**

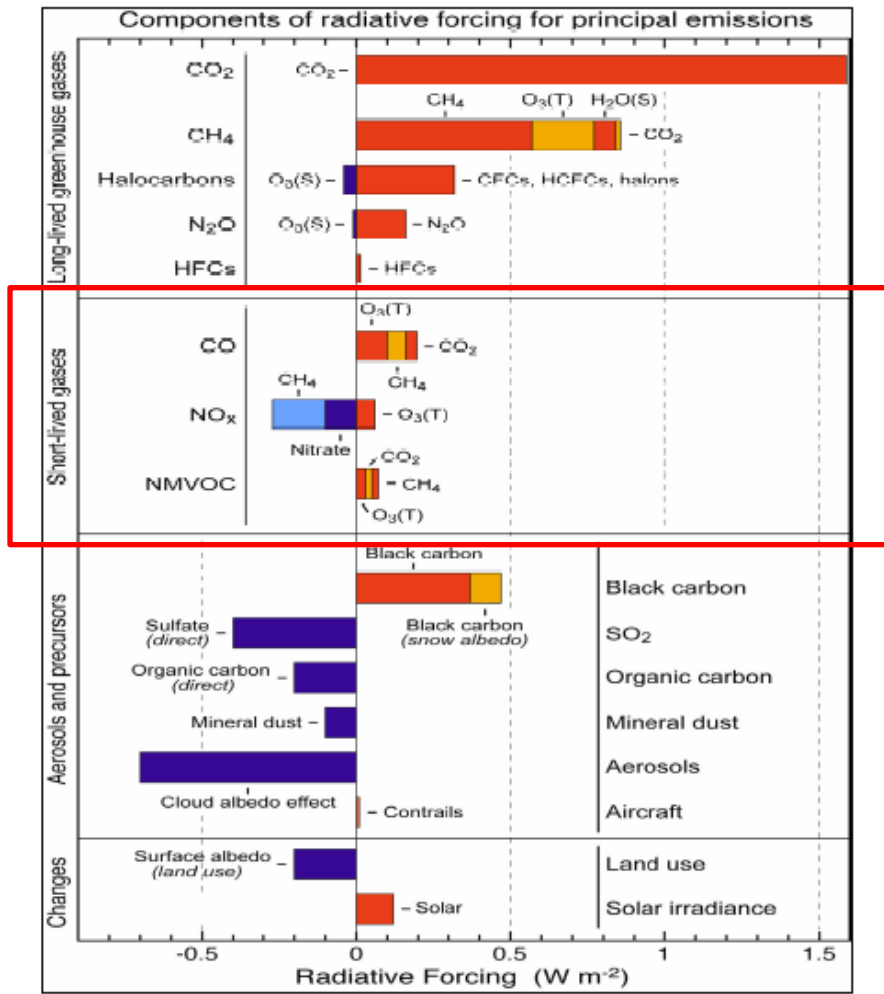
➤ Short-lived Climate Forcers

SLCFs* matter as much as CO2



[IPCC, 2007]

BC responsible for 50% or nearly 1.0° C of the 1.9° C temperature increase in the Arctic from 1890 to 2007 (Shindell, Faluvegi 2009)



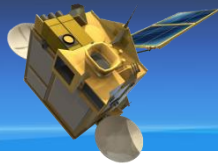
[Witherspoon, EPA SLCF Workshop, 2010]



*Short-lived Climate Forcers

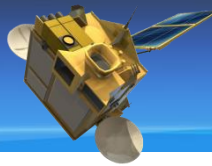


Greenhouse Gases vs Aerosol



Greenhouse gases (GHG)	Aerosols
Greenhouse effect	Whitehouse effect Global dimming
Warming (+)	Cooling (-) sulfate Warming (+) black carbon
global distribution	localized
several to hundreds years	5~7 days
Low Uncertainty	Moderate to High Uncertainty

Greenhouse Gases vs. Aerosol



“There is little disagreement that the rise of aerosols since pre-industrial times has led to both a substantial reduction in solar radiation at the surface and increased solar heating of the atmosphere itself. But global models disagree as to the magnitude of these effects.”

Nine different global climate models	
Greenhouse Gases (GHGs)	$+1.66 \pm 0.17 \text{ W/m}^{-2}$
Aerosols	$-0.22 \pm 0.16 \text{ W/m}^{-2}$ $(+0.04 \sim -0.41 \text{ W/m}^{-2})$

(Seinfeld, *Nature Geoscience*, 2008)

Considering near-term climate impacts as an additional effect of air pollutants

<http://gains.iiasa.ac.at>



- Near-term forcing of air pollutants
 - Warming: BC, O₃ (i.e., CH₄, CO, NO_x, VOC)
 - Cooling: SO₂, OC
 - accelerates or delays ongoing climate change at the regional scale,
 - changes regional weather circulation and precipitation patterns.
- Accelerated melting of Arctic ice and glaciers through deposition of (black) carbon

could be considered as an additional effect of air pollutants
(on top of their health and ecosystems impacts)

(M. Ammann, 2010)

SLCF

- Health impacts of SLCF are important and immediate
 - Should be integral element of climate policy considerations
- Black carbon reductions would benefit both health and climate
 - Directionality is known even if magnitude is uncertain
 - Of the ways to improve PM air quality, reductions in black carbon are likely the best for climate
- O₃ precursor reductions would also benefit health and climate
 - Of the ways to improve ozone air quality, reducing methane, CO, VOCs (in that order) are likely best for climate
- Eventual strategies will need to consider health, climate, and other effects.
 - Need integrated management view



Transboundary pollution in East Asia



Long-range transboundary transport of air pollutants in Northeast Asia (source-receptor regions)

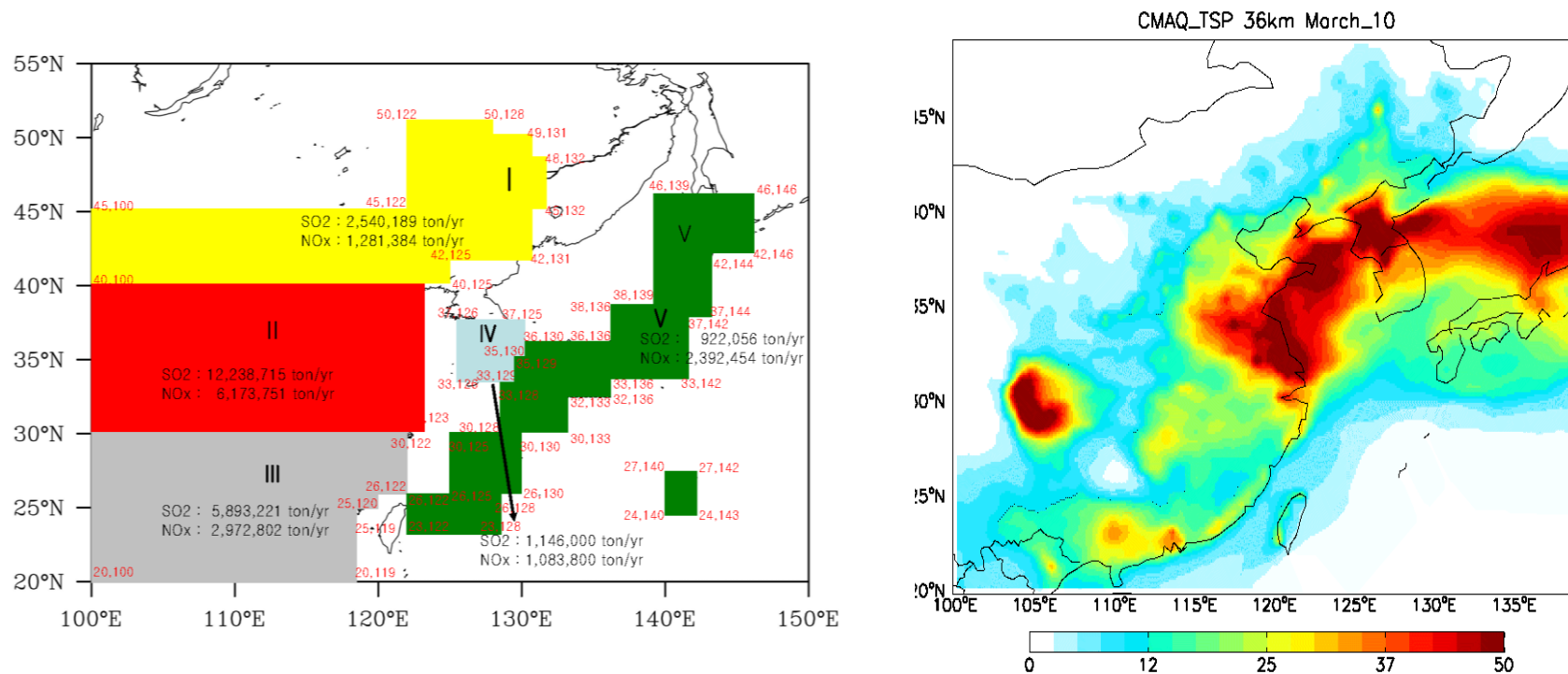


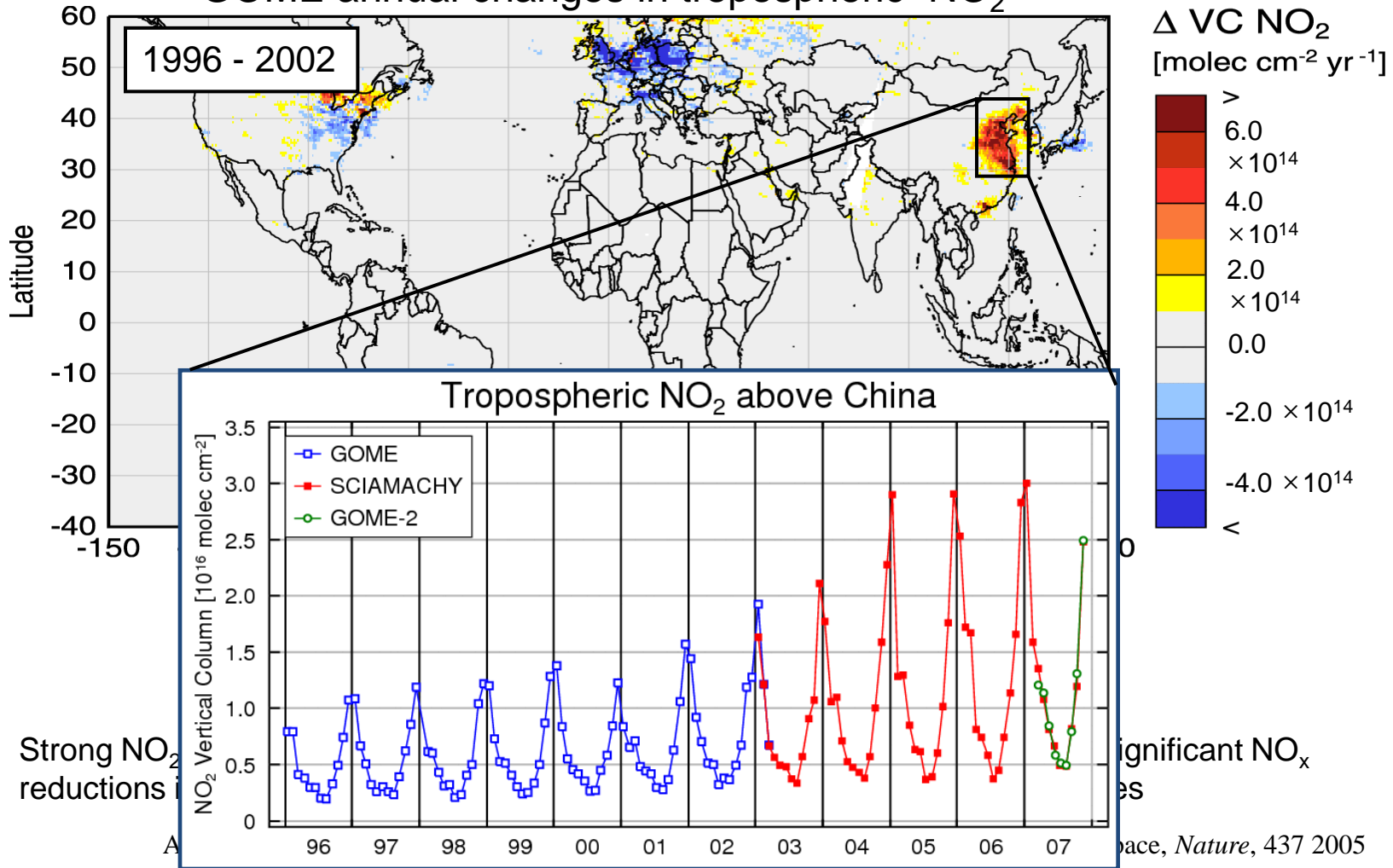
Fig. 8. Total emission amounts of SO₂ and NO_x for each source/receptor region.

[LTP project, 2007]

Satellite Retrieval of Tropospheric NO₂ column



GOME annual changes in tropospheric NO₂



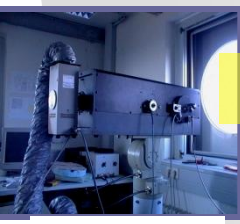
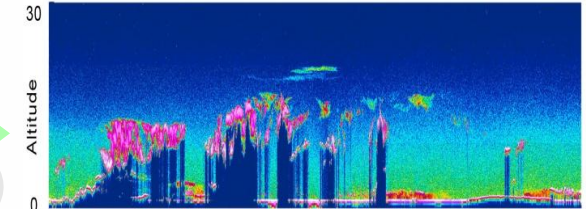
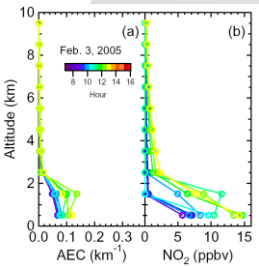
Integrated Diagnostics of Atmospheric Environment



Satellite (MODIS, GOME
SCIAMACHY, OMI, GEMS etc)



Aircraft
monitoring
(AMAX-DOAS)



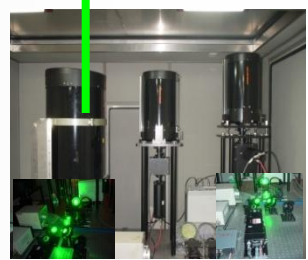
LP-DOAS



MAX-DOAS or
Imaging-DOAS



Mobile lab



Lidar



Sunphotometer



In-situ:
Semi-continuous
**Chemical, Physical,
Optical**
properties of aerosol

- Transmissometer
- Nephelometer
- Aethelometer
- PILS-IC/TOC
- Aerosol Spectrometer
- Gas monitor



Differential Optical Absorption Spectroscopy (DOAS)

- DOAS technique is 30 years old ...
- Many applications have been demonstrated and used, but research is still ongoing on many aspects

Active long-path DOAS



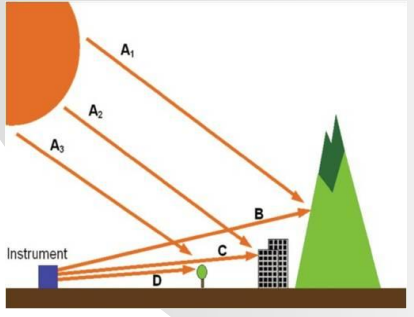
Passive MAX-DOAS



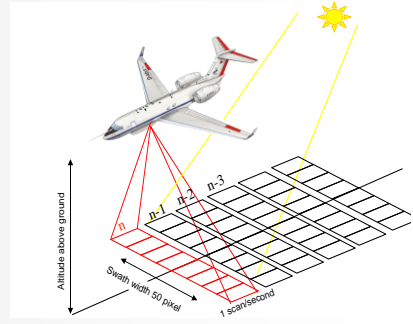
Passive Imaging DOAS



Passive ToTal*-DOAS



Airborne Multi-Axis DOAS



DOAS from space
(GOME, SCIAMACHY, OMI, and GEMS)



*Topographic Target Light scattering



DOAS principle

Remove by high-pass filtering

Lambert-Beer's Law:

$$I(\lambda) = I_0(\lambda) \cdot e^{-[\underbrace{\sum \sigma'_i(\lambda) \cdot c_i \cdot L}_{\text{narrow-}} + \underbrace{(\sigma_{bi} \cdot c_i + \epsilon_{\text{Ray}}(\lambda) + \epsilon_{\text{Mie}}(\lambda)) \cdot L}_{\text{wide band extinction}}] \cdot T(\lambda)}$$

Rayleigh scattering
~ λ^{-4}

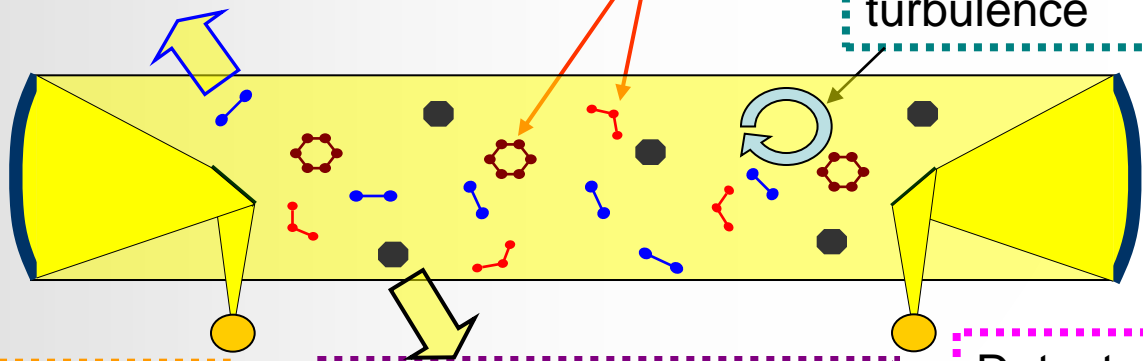
Trace gas absorption
absorption cross section $\sigma_i(\lambda)$

turbulence

Lamp $I_0(\lambda)$

Mie Scattering
~ $\lambda^{-(1...3)}$

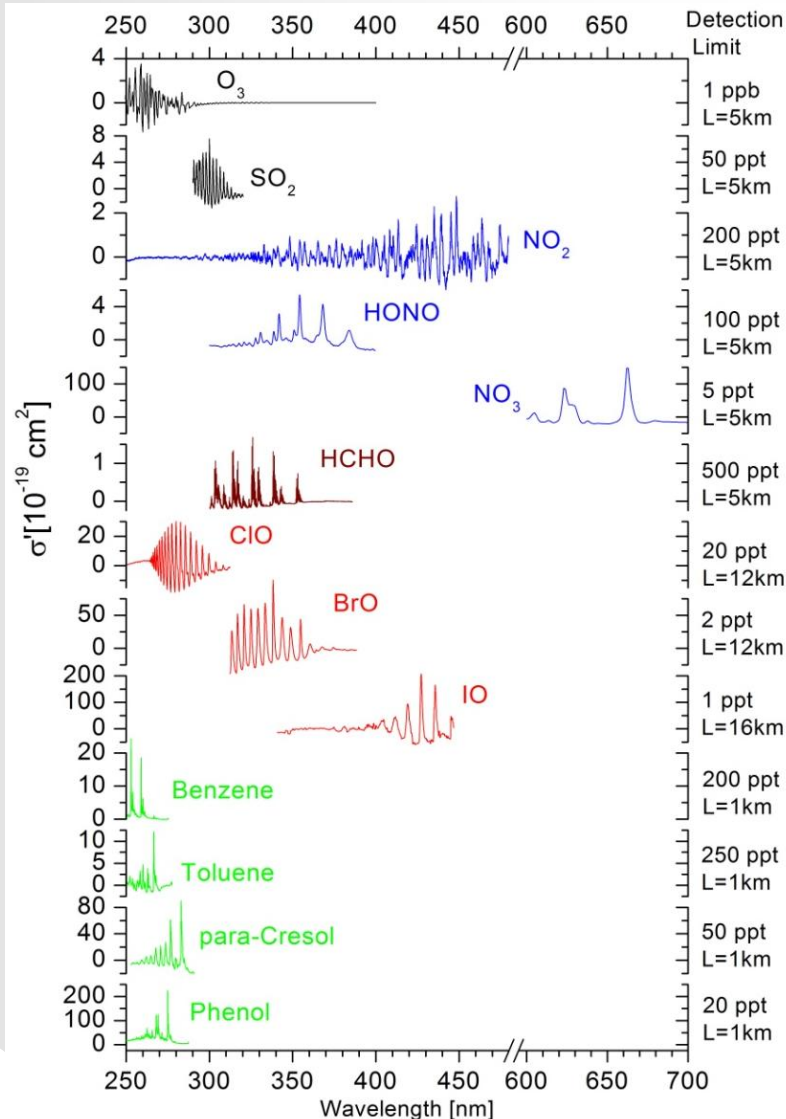
Detector $I(\lambda)$



[Platt, 1994]



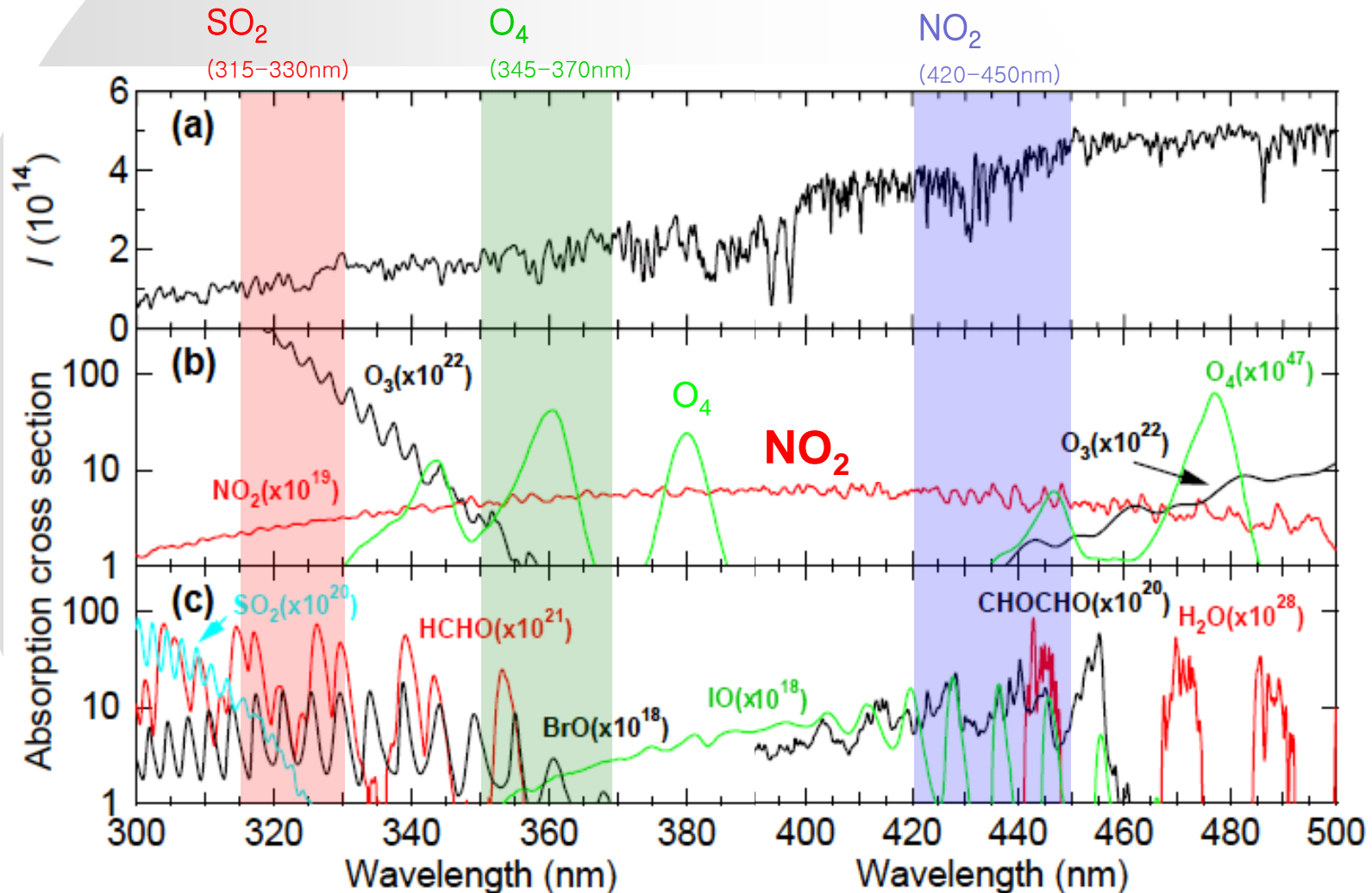
Absorption cross-sections in UV-Vis range



[IUP, Univ. Heidelberg, Germany
(<http://www.iup.uni-heidelberg.de>)]



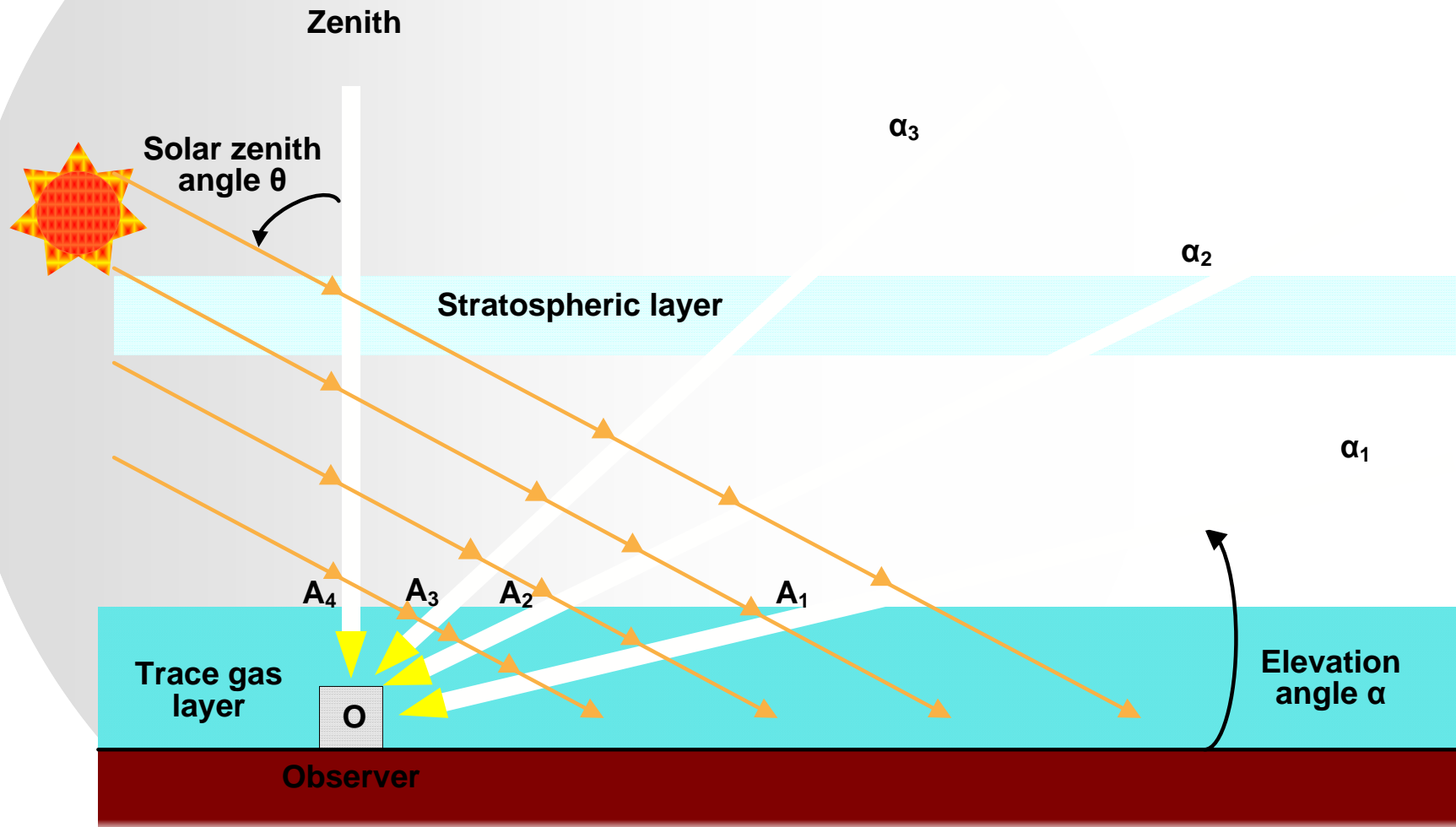
Absorption cross-sections in UV-Vis range



(Courtesy of H. Irie)



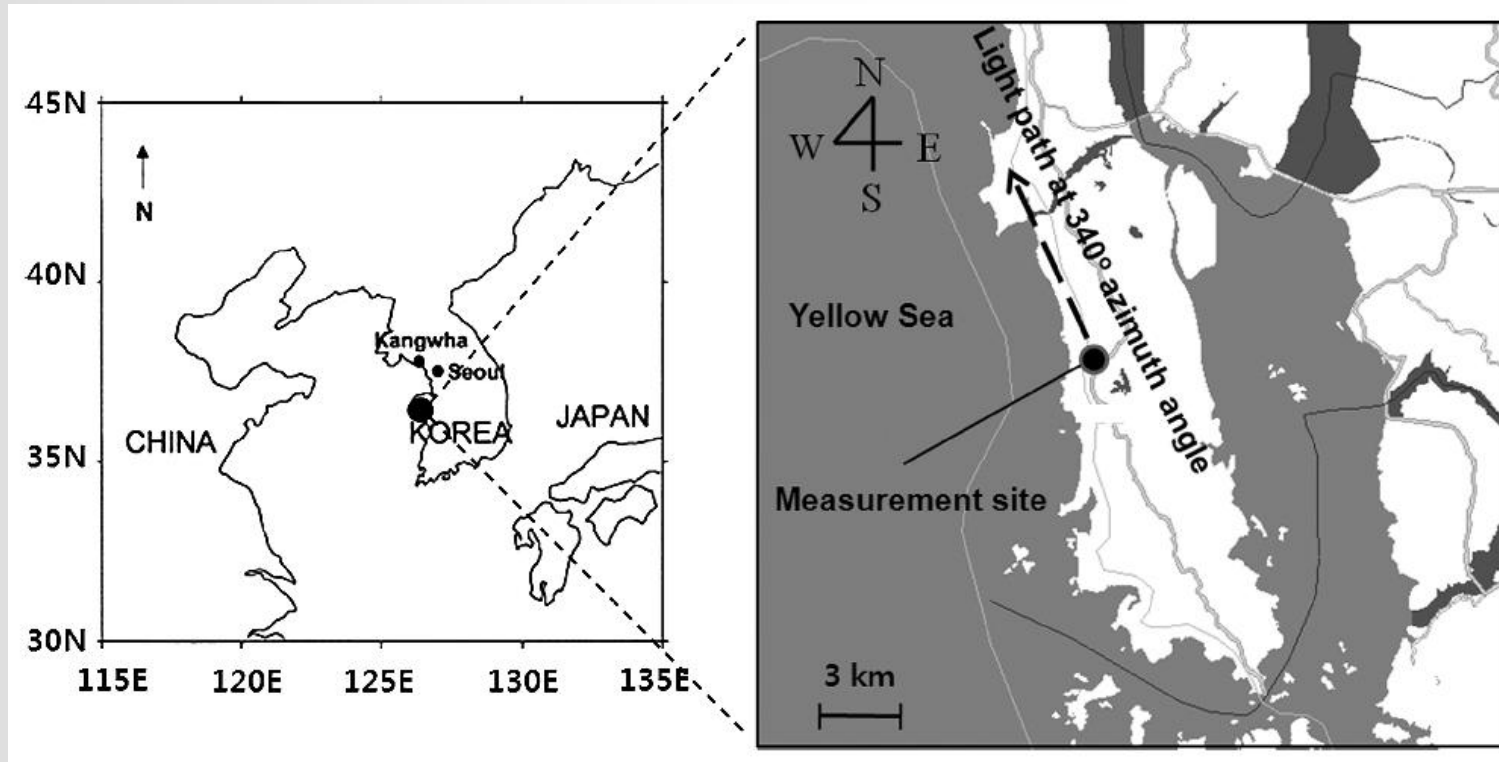
Multi-Axis DOAS (MAX-DOAS) viewing geometry



[Hoeningner, 2002]



I . MAX-DOAS measurements of ClO, SO₂ and NO₂ at Anmyeon Island (Korea GAW site)



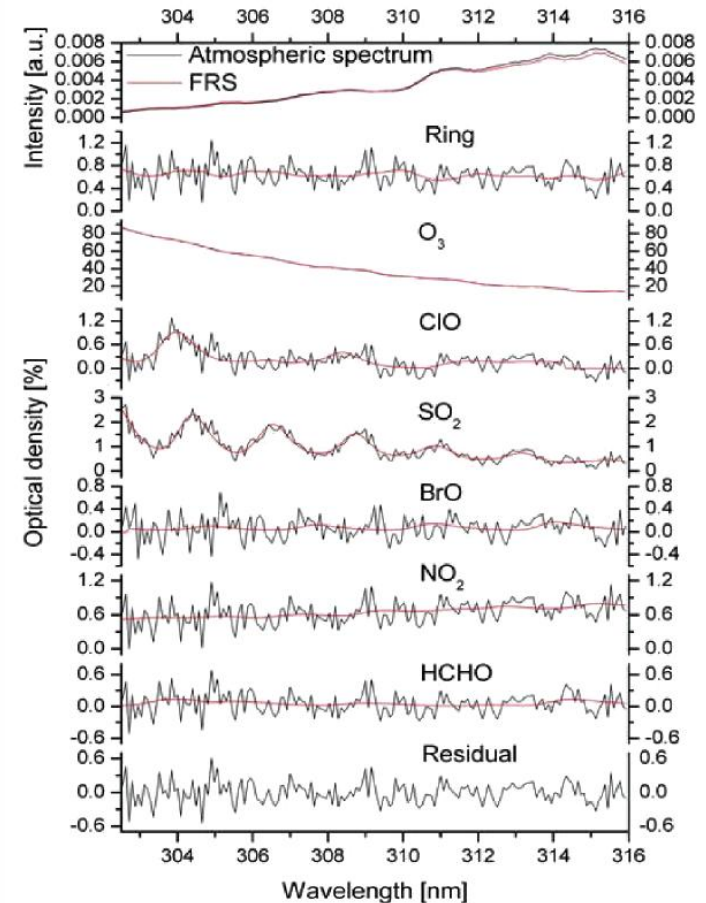
Site: The Korea Global Atmosphere Watch Observatory (KGAWO)
(36.54° N, 127.12° E), Anmyeon Island
Period: 27 May ~ 9 June 2005.

[Lee, C. et al., 2008]

I . Analysis

Specifications for the MAX-DOAS spectrum evaluation for ClO, NO₂, and SO₂

Molecule	Wavelength range, nm	Polynomial order	Cross-sections included in the fitting procedure
ClO	302.5–316	3	ClO, BrO, SO ₂ , HCHO, NO ₂ , O ₃ , Ring, FRS
SO ₂	303.5–316	3	ClO, SO ₂ , BrO, NO ₂ , O ₃ , Ring, FRS
NO ₂	399–418	3	NO ₂ , O ₄ , O ₃ , Ring, FRS

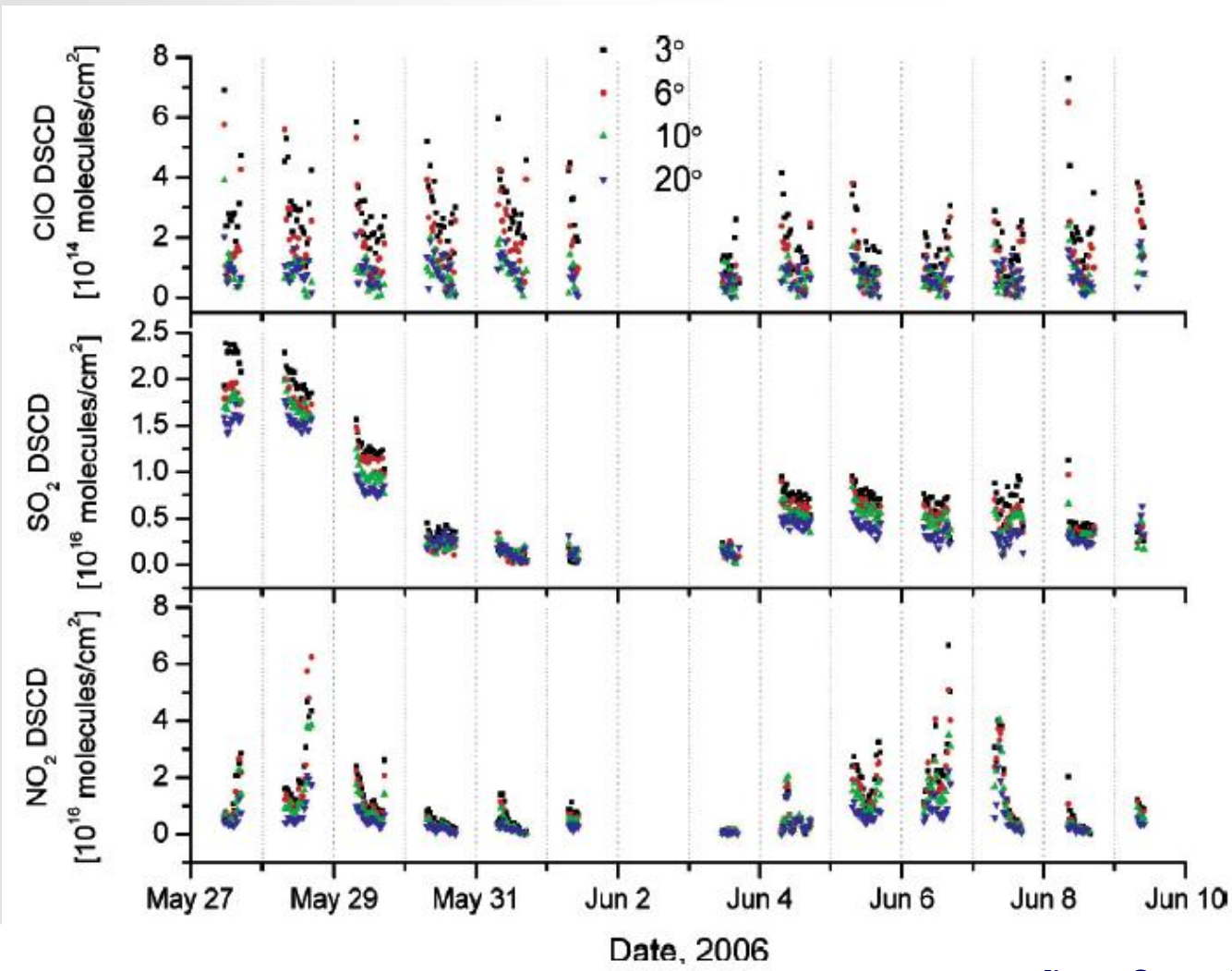


An evaluation example of a ClO SCD from the MAX-DOAS spectrum taken at a 3° elevation angle at 16:12 on 5 June 2005 (LT)

[Lee, C. et al., 2008]



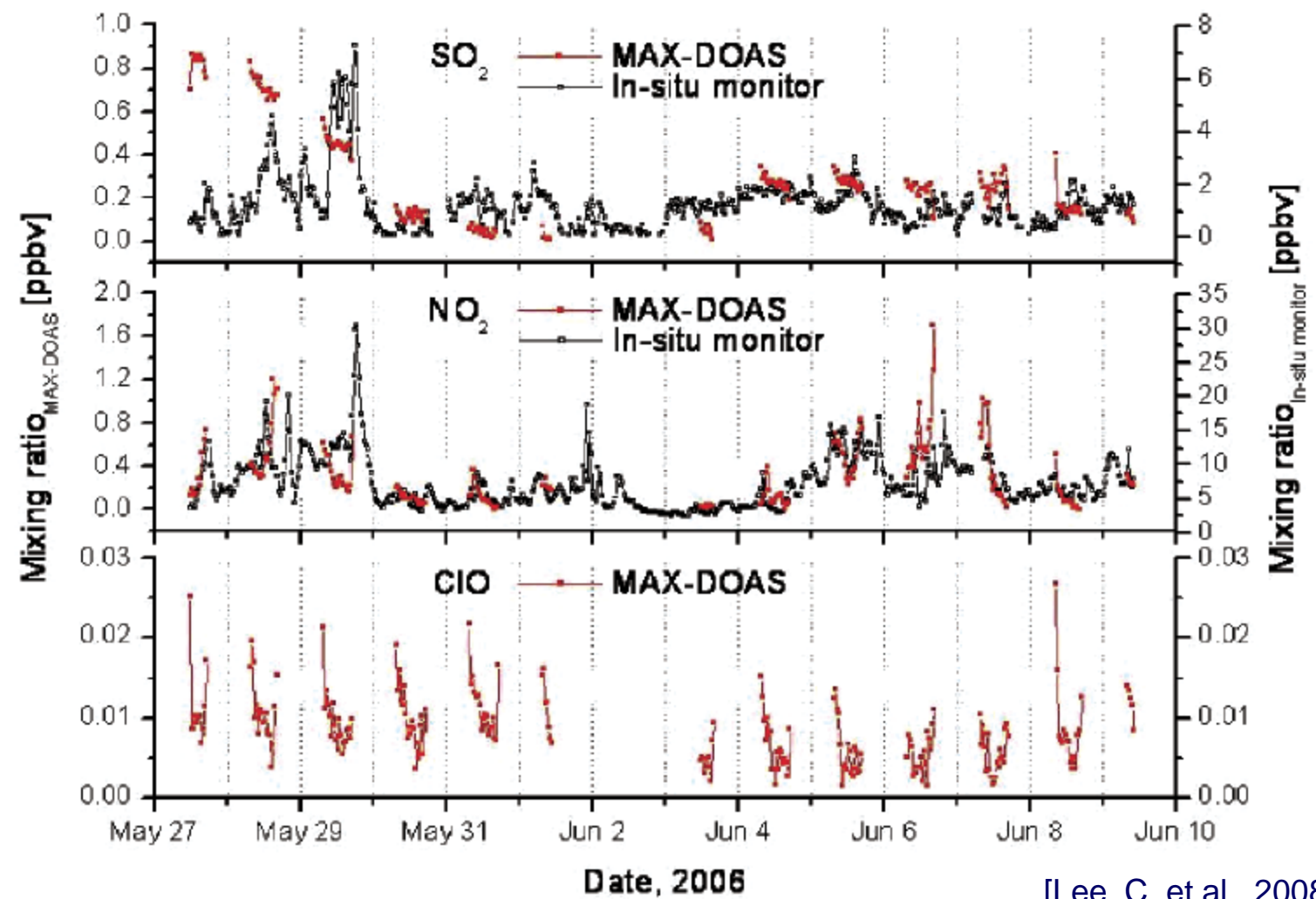
I . ClO, SO₂ and NO₂ differential slant column densities (DSCD)



[Lee, C. et al., 2008]



I . Mixing ratios of SO₂, NO₂, and ClO determined by the MAX-DOAS and in situ monitors.



[Lee, C. et al., 2008]

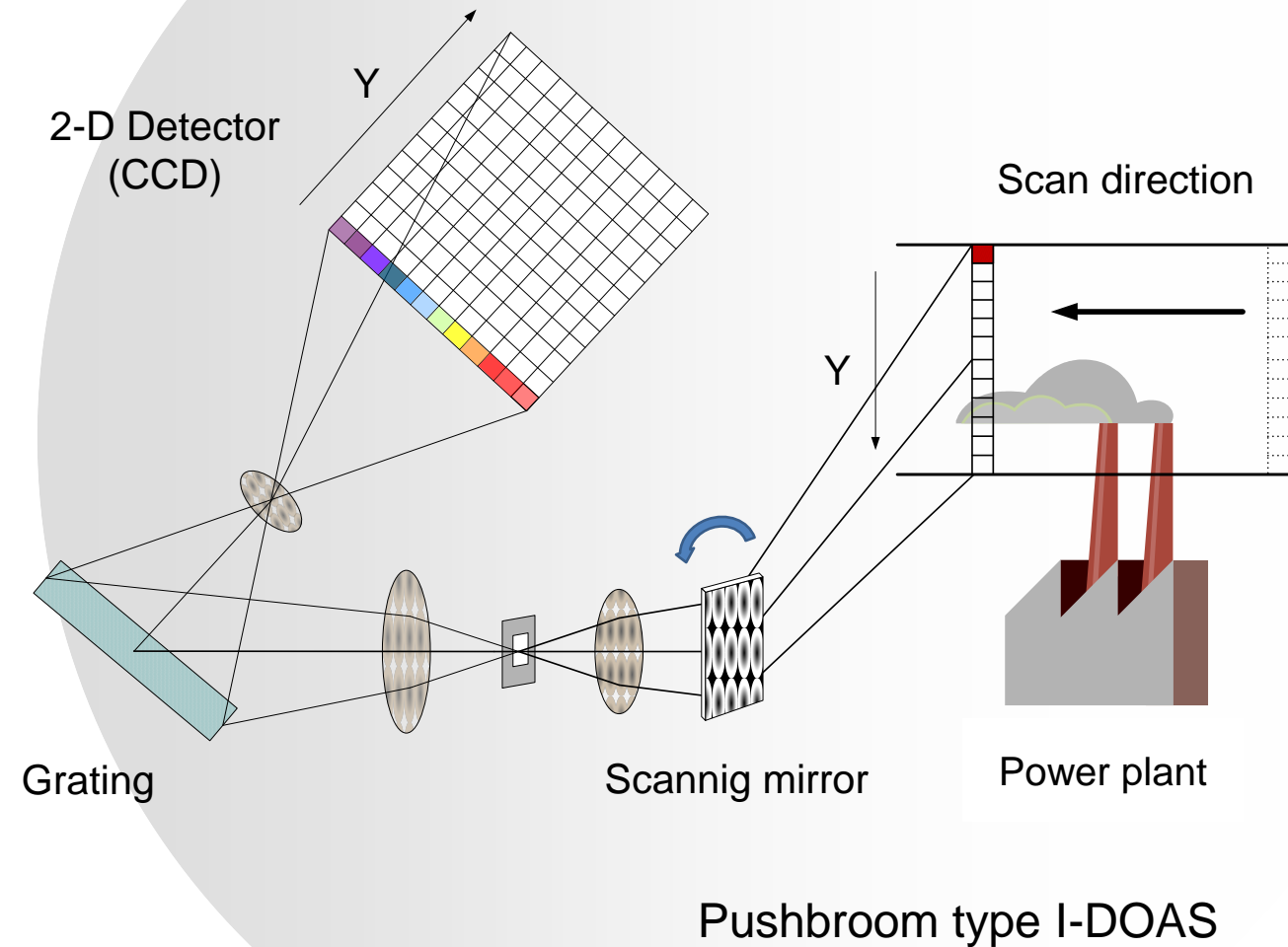


I . Summary

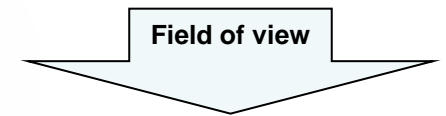
- ClO, SO₂ and NO₂ have been directly observed in the mid-latitude coastal boundary layer by the MAX-DOAS technique.
- The diurnal behavior of ClO was consistent with a source based on the heterogeneous processing of sea-salt aerosol. (ClO level was as high as ~27 pptv (with a mean of 8.4 pptv)
- Mean SO₂ and NO₂ levels observed during the measurement period were 296 (±233) and 305 (±284) ppbv, respectively.
- High SO₂ and NO₂ concentrations measured during the two event periods might have an effect on atmospheric halogen chemistry through reactive halogen release processes by the attack of strong acids on sea-salt aerosols, and through reducing processes by the reaction of ClO with NO₂.



Imaging DOAS viewing geometry



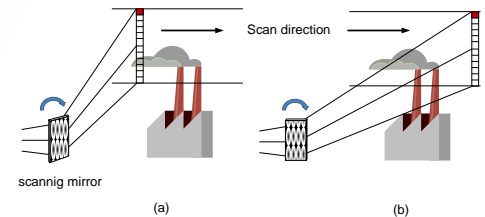
Slant line scanning



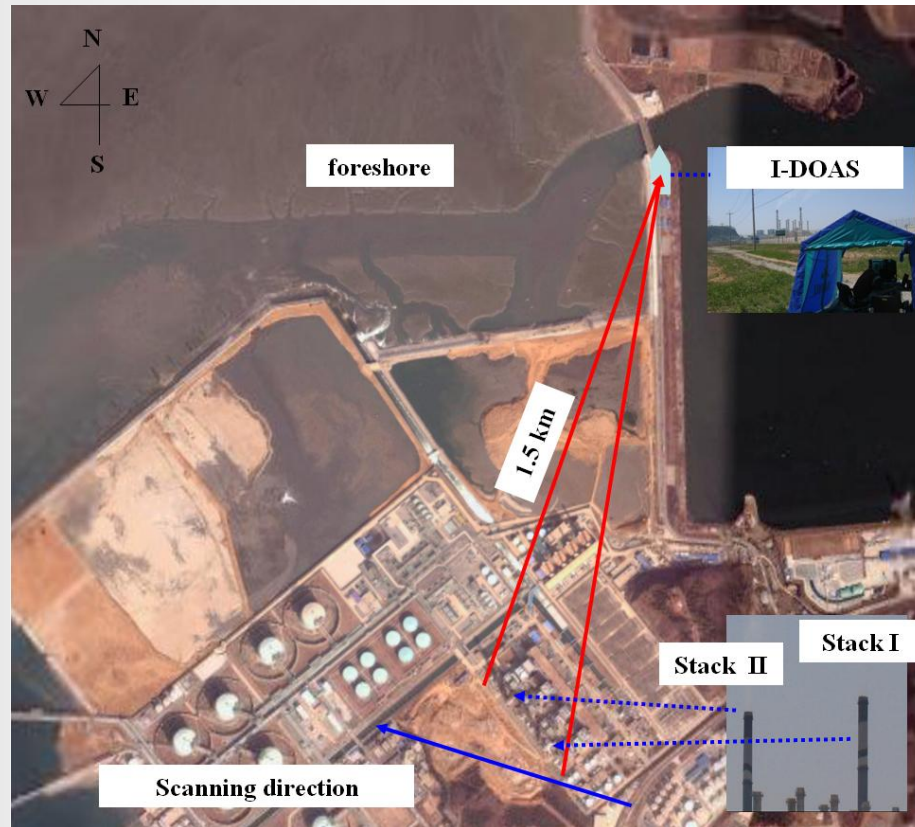
Superficial scanning



Volume scanning (Scanning mirror)



II. Imaging DOAS measurements of NO₂



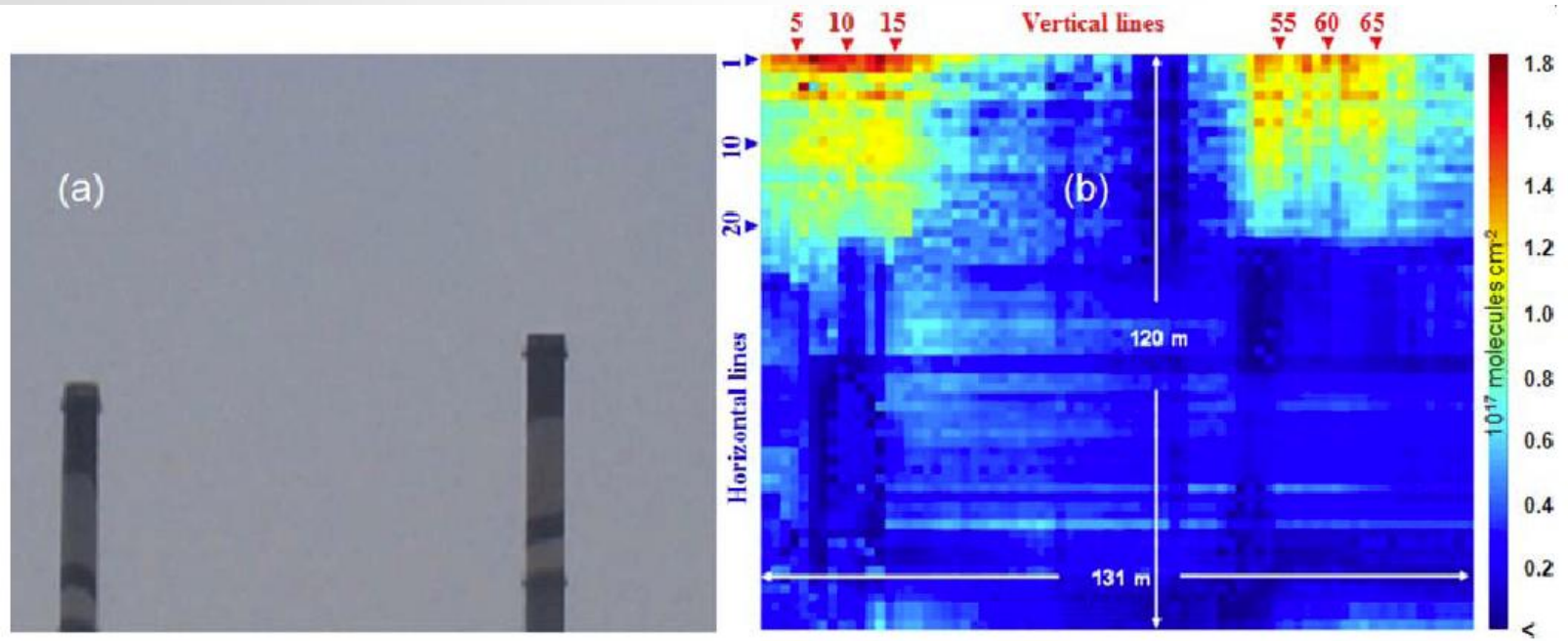
Site: the rooftop of a local agricultural administration facility at a distance of 1.7 km north from the Pyeongtaek thermal power plant (37.00° N, 126.47° E)

Period: 15 September, 2005

[Lee, H. et al., 2009]

II. NO₂ distribution in the power plant plume

The measurement was taken 1.7 km north of the stacks of the Pyeongtaek Power Plant between 9:15 A.M. and 9:37 A.M. on 15 September 2005



(a) is invisible in the RGB color photograph

(b) is visualization of the distribution of NO₂

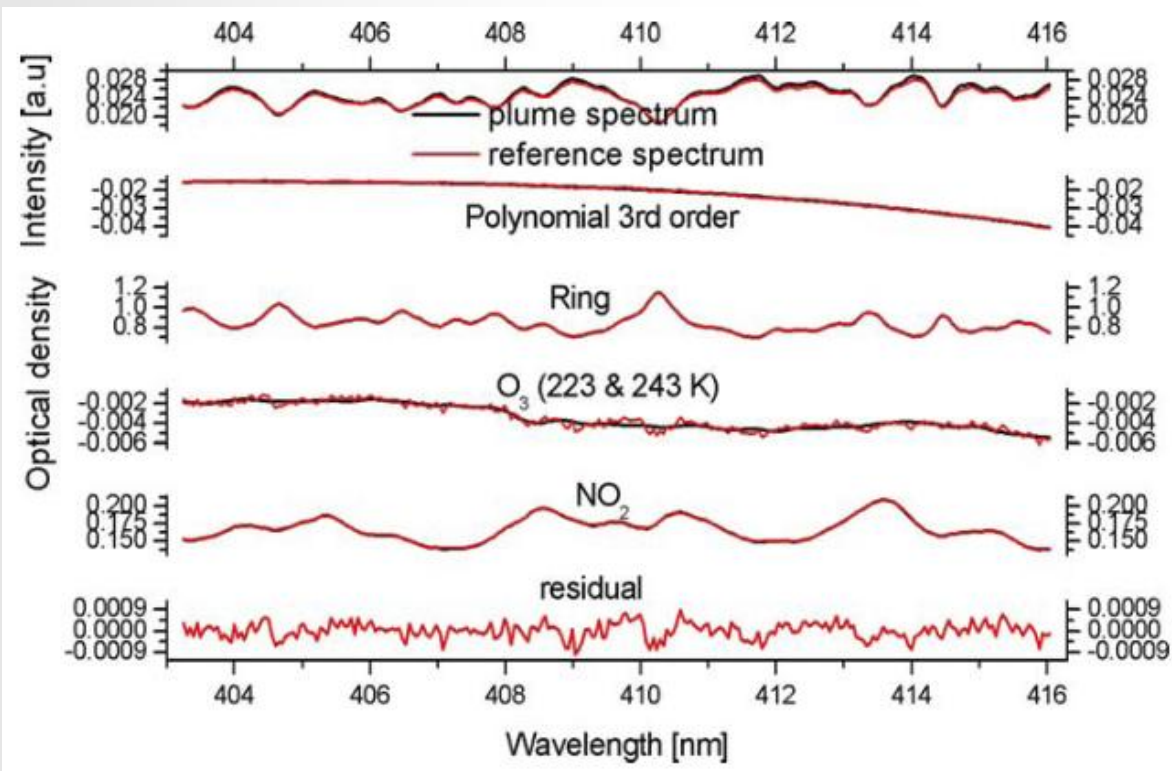
-Background SCD
: ~0.2 × 10¹⁷ molecules cm⁻²

-Max. SCD observed 45 m above the 1st stack exit
: 1.8 × 10¹⁷ molecules cm⁻²

[Lee, H. et al., 2009]



II. Analysis

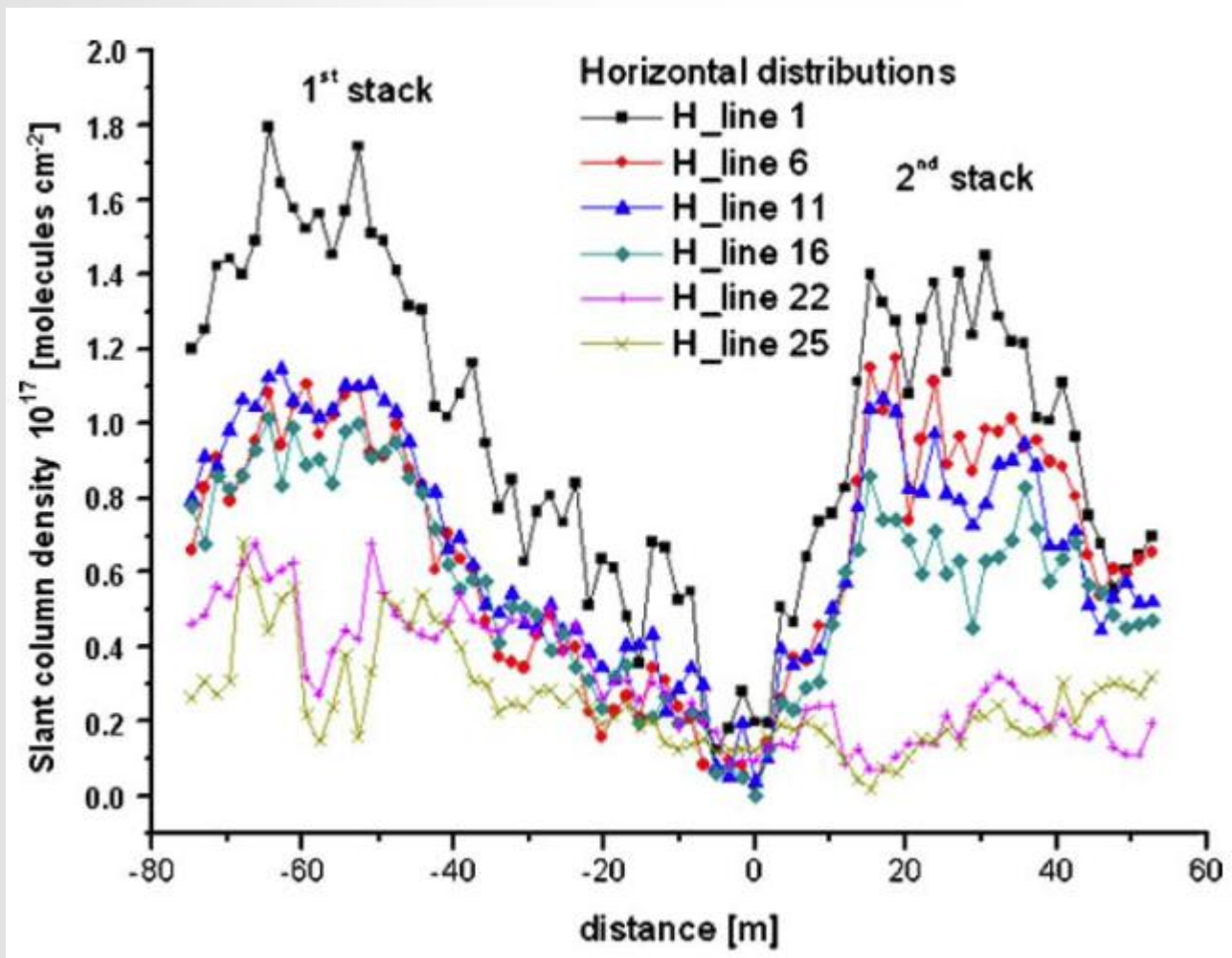


Molecule	Wavelength range (bands ^a)	Polynomial order	Cross-sections included in fitting routine
NO ₂	309.5 - 417 nm	3	NO ₂ , O ₃ , FRS, Ring

[Lee, H. et al., 2009]



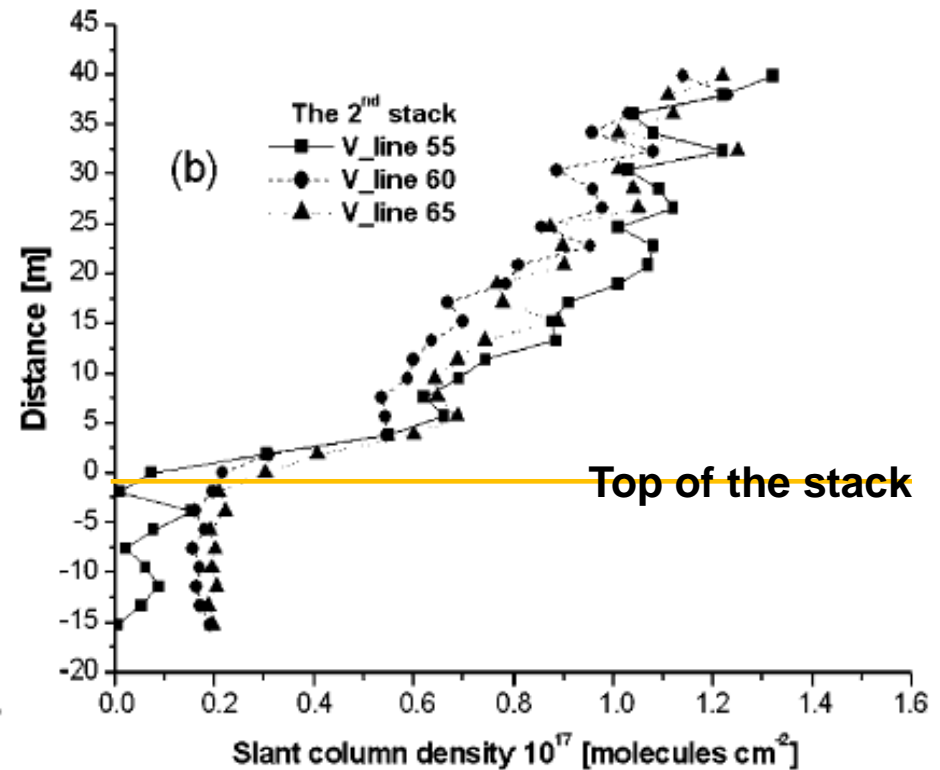
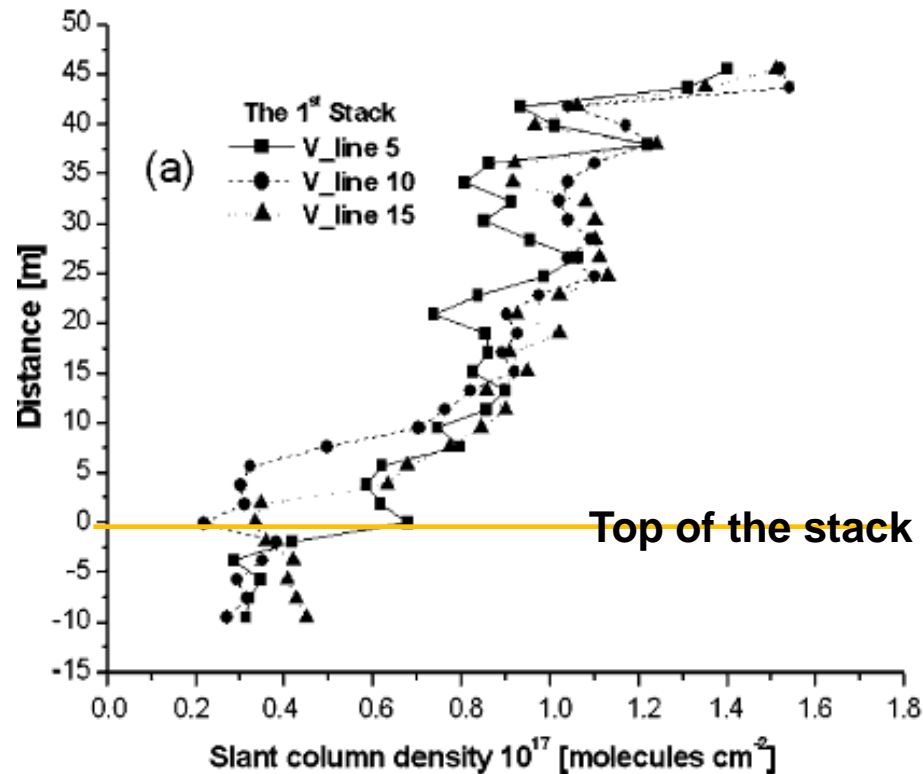
II. Horizontal NO₂ slant column density (SCD)



[Lee, H. et al., 2009]



II. Vertical NO_2 slant column density distributions above stacks



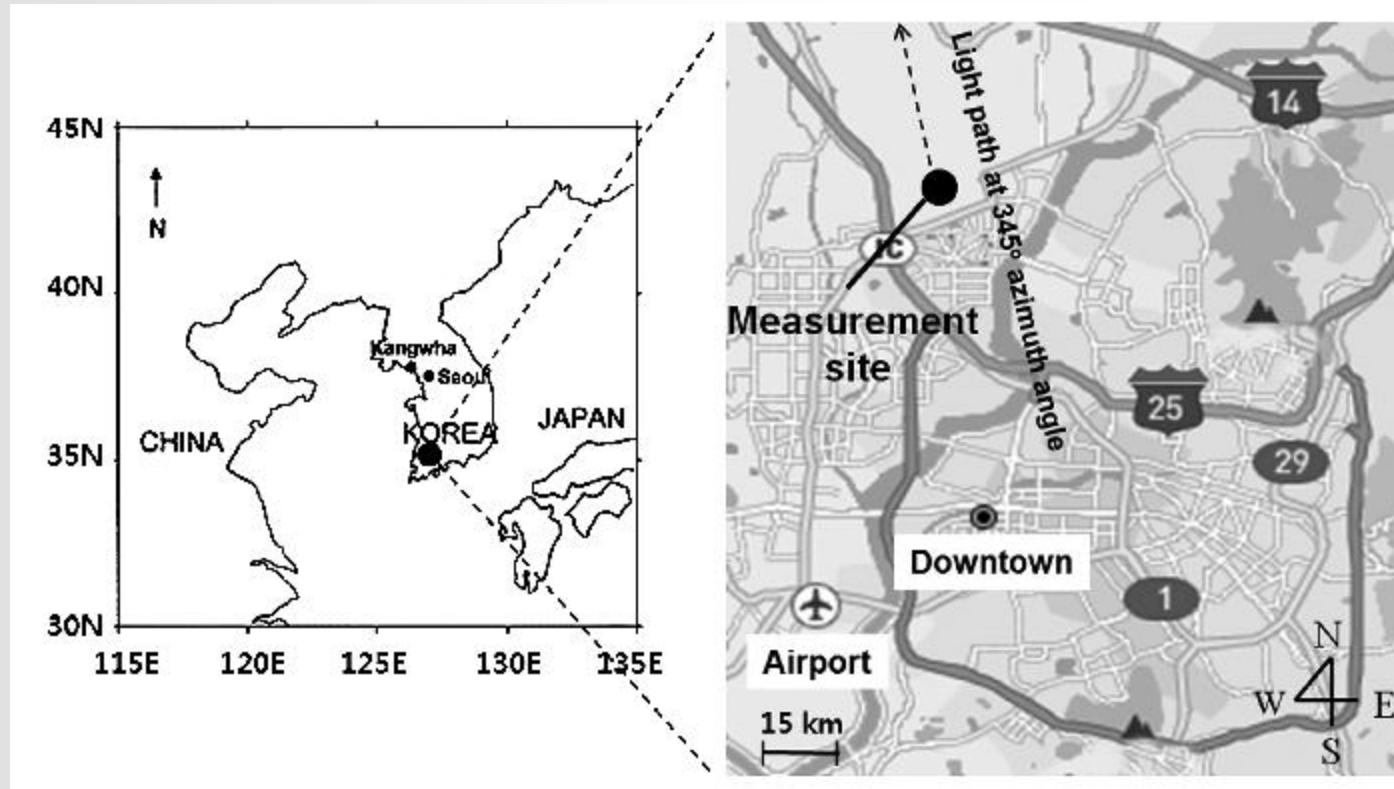
[Lee, H. et al., 2009]

II. Summary

- This study is the first to measure the vertical NO₂ gradient in the 40–45 m above the exits of power plant stacks with a high spatial resolution of 1.9 (vertical)×1.7 m (horizontal).
- Rates of NO₂ increase within two rising plumes were directly obtained using the I-DOAS technique, revealing rates of 60 and 70 ppb s⁻¹, corresponding to 6 and 7 ppb m⁻¹, respectively.
- The obtained rates of NO₂ increase are only applicable to areas close to the stack.



III. MAX-DOAS measurement of Aerosol

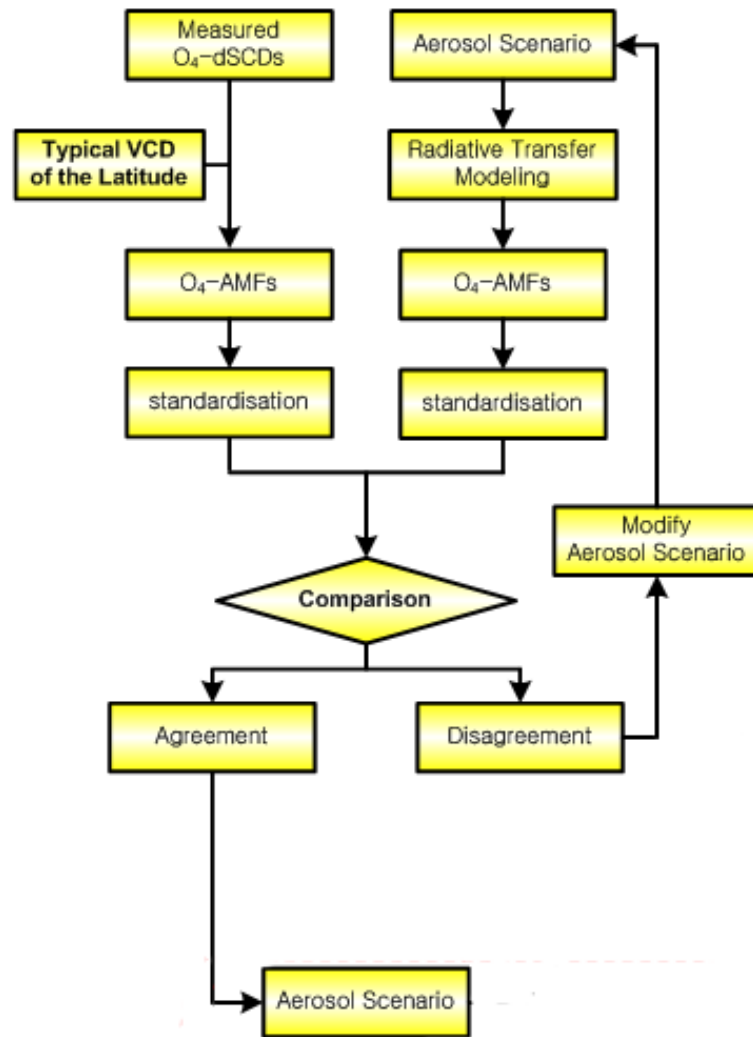


Site: Gwangju (35.11 N, 126.54E)

Period: 27 February - 18 May 2008 ,
a period that includes two severe Asian dust storm days

[Lee, H. et al., 2009]

III. Aerosol Retrieval Algorithm



1. Obtain O_4 DSCDs from MAX-DOAS measurement data
2. Calculate O_4 AMFs with known vertical O_4 column density at various elevation angles.
3. Vary aerosol profile as an input for RTM until simulated O_4 matches with those observed at all elevation angles.
4. An optimized aerosol profile is obtained from the agreement for all elevation angles.

[Sinreich et al., 2005]

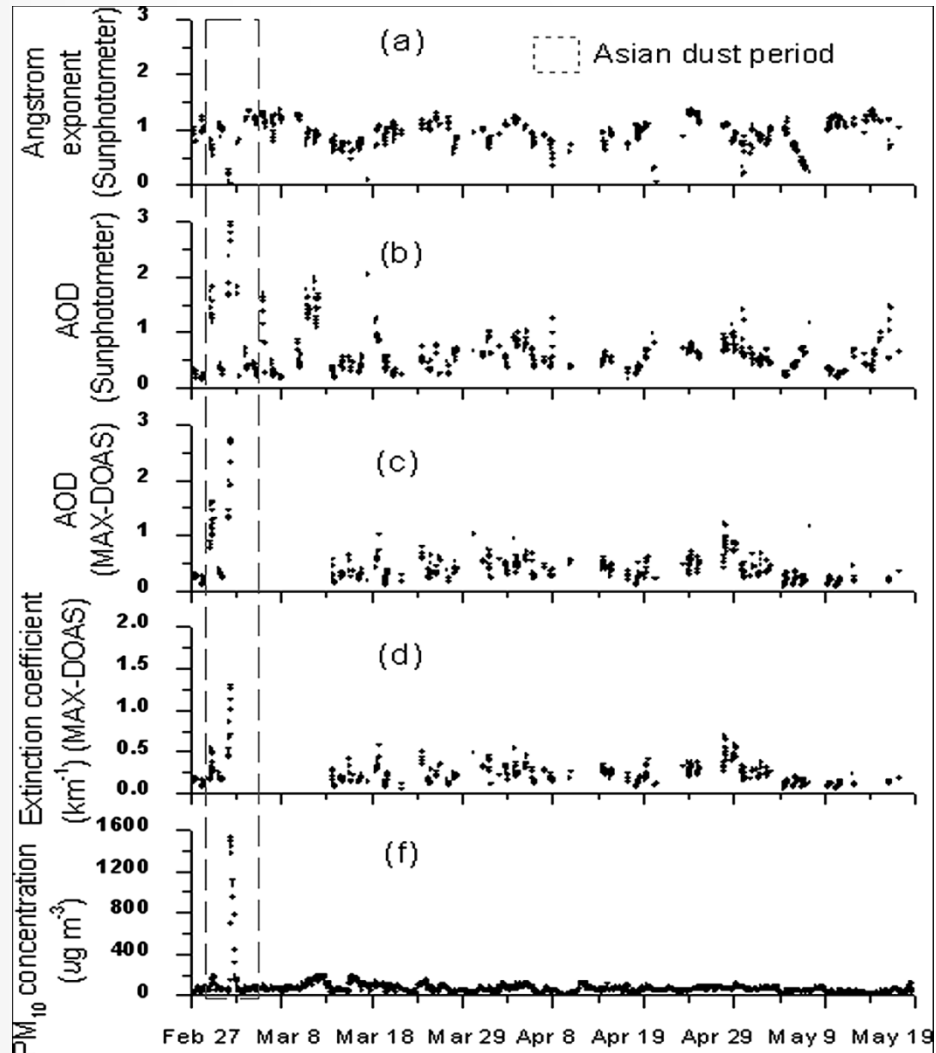


III. Temporal variations of measured aerosol quantities

as low as 0.54 on 29 February
0.01 on 2 March

Note: Low A° exponent values on those two days suggest that the aerosol mostly consisted of coarse particles

PM10 concentrations range from 37 to 1521 $\mu\text{g m}^{-3}$ on 2 March
from 45 to 185 $\mu\text{g m}^{-3}$ on 29 February

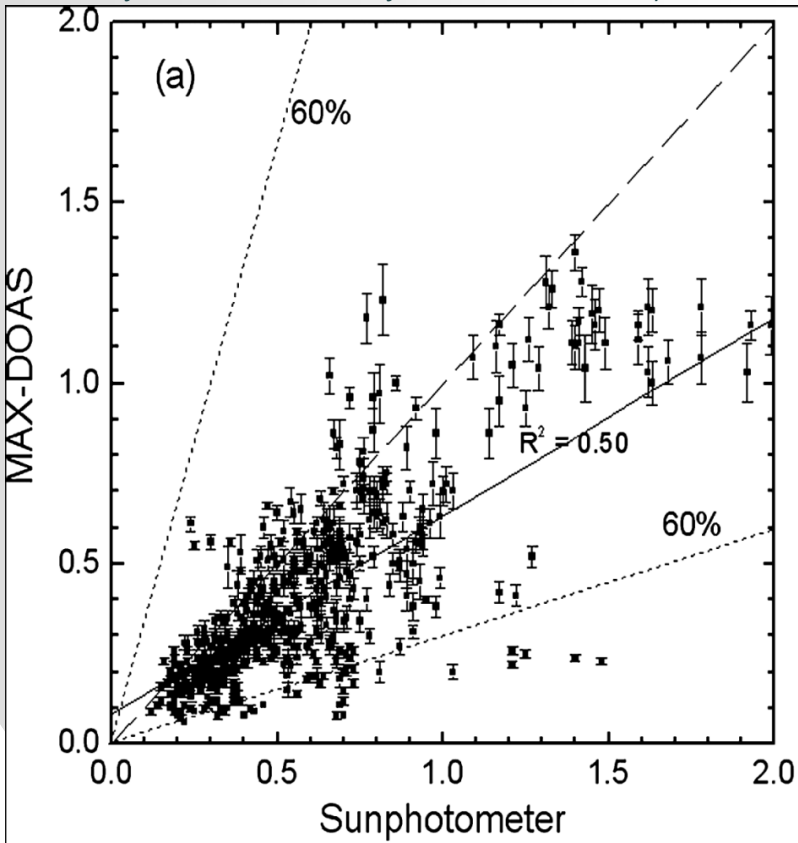


[Lee, H. et al., 2009]

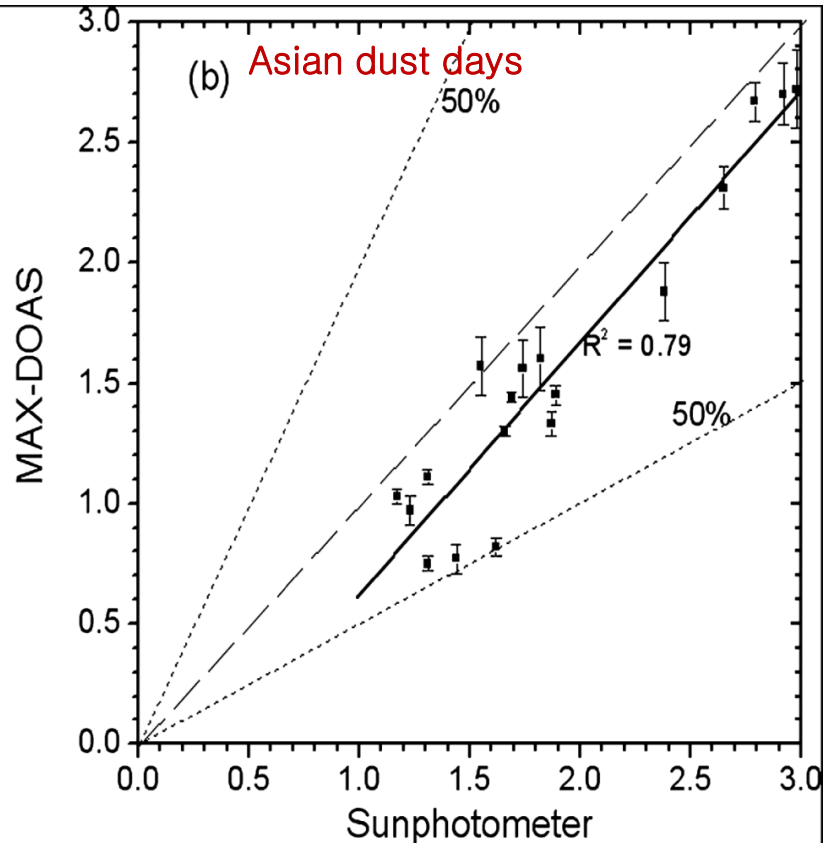


III. MAX-DOAS vs. Sunphotometer

(27 February–18 May 2008, except the two Asian dust days on 29 February 29 and 2 March)



(on 29 February 29 and 2 March)



[Lee, H. et al., 2009]



III. Summary

- Retrieved AODs and AECs at the 0–1 km layer were validated by comparison with collocated sunphotometer data and surface PM10 concentrations measured by a beta gauge sampler.
- Scatter of the correlation is within 50% on the two Asian dust days.
- Much less correlation was also observed between the surface PM10 and AEC at 0.5 km above ground level on the Asian dust days than that obtained during the non episodic days.
- As investigated in recent studies (Irie et al. 2008a, 2009; Lee et al. 2009), several factors can cause uncertainties involved in MAX-DOAS measurements including a priori values, constants such as single scattering albedo, and fixed surface albedo, as well as atmospheric conditions such as fogs and clouds at lower altitudes.



Summary (1)

- Optical remote sensing techniques have been preferred for measurements of atmospheric trace gases and aerosol.
- **DOAS** has been used as a powerful tool for ground-based optical remote sensing of atmospheric **trace species** (e.g., SO₂, NO₂, O₃, HCHO, BrO, ClO, and VOCs) and aerosol.
- The more recently developed **Imaging DOAS** technique provides information on the spatially resolved two dimensional distributions of trace gases.
- DOAS technique has been applied as an alternative tool for the detection of **atmospheric aerosols**.
- The **satellite** remote sensing approach associated with the spectral DOAS fit technique has been successfully employed for the measurements of tropospheric trace gases on **global and regional scales**.



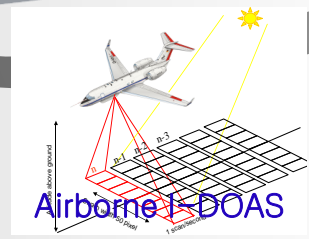
Summary (2)

Optical remote sensing of atmospheric trace gases and aerosol

Ground-based Max-DOAS



Airborne MAX-DOAS or Imaging-DOAS



Satellite observations



Thank you!

E-mail: yjkim@gist.ac.kr



IV. Major Satellite Sensors for the Trace Gas Measurements

TOMS
(1978 – 2008)

- ❖ **T**otal **O**zone **M**apping **S**pectrometer
 - onboard Nibus, Adeos, Earth Probe
 - O₃ (additionally SO₂)



GOME
(1995 – 2003)

- ❖ **G**lobal **O**zone **M**onitoring **E**xperiment
 - onboard ERS-2
 - O₃, NO₂, OCIO, BrO, HCHO, SO₂, H₂O, etc.



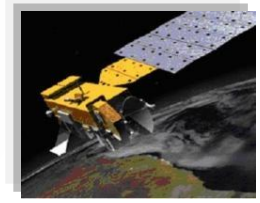
SCIAMACHY
(2002 –)

- ❖ **S**Canning **I**maging **A**bsorption spectro**M**eter for **A**tmospheric **C**Hartograph**Y**
 - onboard ENVISAT
 - O₃, NO₂, OCIO, BrO, HCHO, SO₂, H₂O, CO, CO



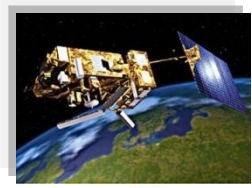
OMI
(2004 –)

- ❖ **O**zone **M**onitoring **I**nstrument
 - onboard EOS-Aura
 - O₃, NO₂, OCIO, BrO, HCHO, SO₂, H₂O, etc.



GOME-2
(2006 –)

- ❖ **G**OME-2
 - onboard MetOp
 - O₃, NO₂, OCIO, BrO, HCHO, SO₂, H₂O, etc.



IV. Major satellite instruments for remote sensing

Instrument	Satellite platform	Measurement period	Equator crossing time (local time)	Spectral range (μm)	Spectral resolution (nm)	Spatial resolution, Nadir (km)	Global coverage (days)
GOME ^a	ERS-2	1995–2003 ^b	10:30	0.23–0.79	0.2–0.4	320 × 40	3
SCIAMACHY ^c	Envisat	2002	10:00	0.23–2.3	0.25–0.4	60 × 30	6
OMI ^d	EOS-Aura	2004	13:30	0.27–0.50	0.5	24 × 13	1
GOME-2	MetOp	2006	09:30	0.24–0.79	0.26–0.51	80 × 40	1.5

^aGlobal Ozone Monitoring Experiment.

^bOperating at reduced coverage since June 2003.

^cSCanning Imaging Absorption SpectroMeter for Atmospheric CHartography.

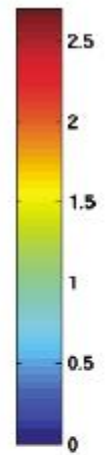
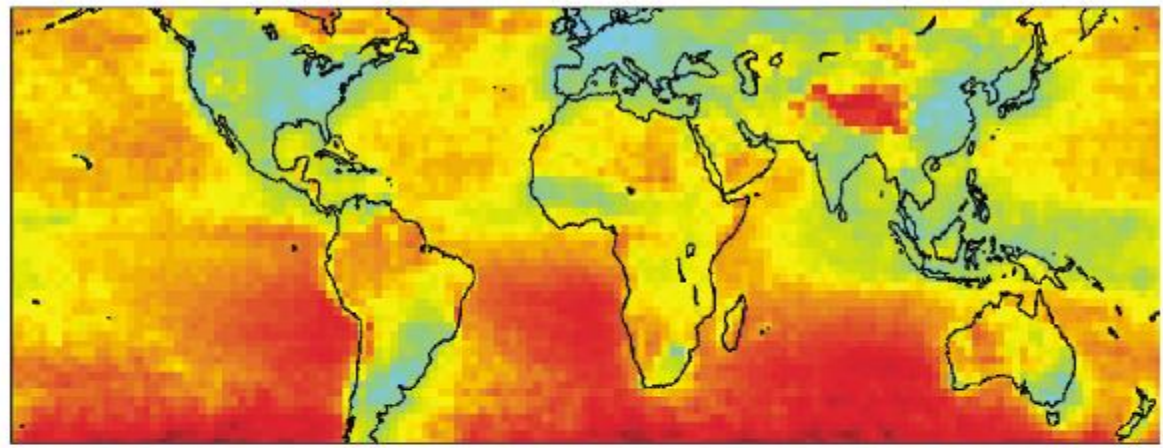
^dOzone Monitoring Instrument.

[Lee, C. et al., 2009]



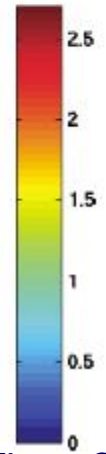
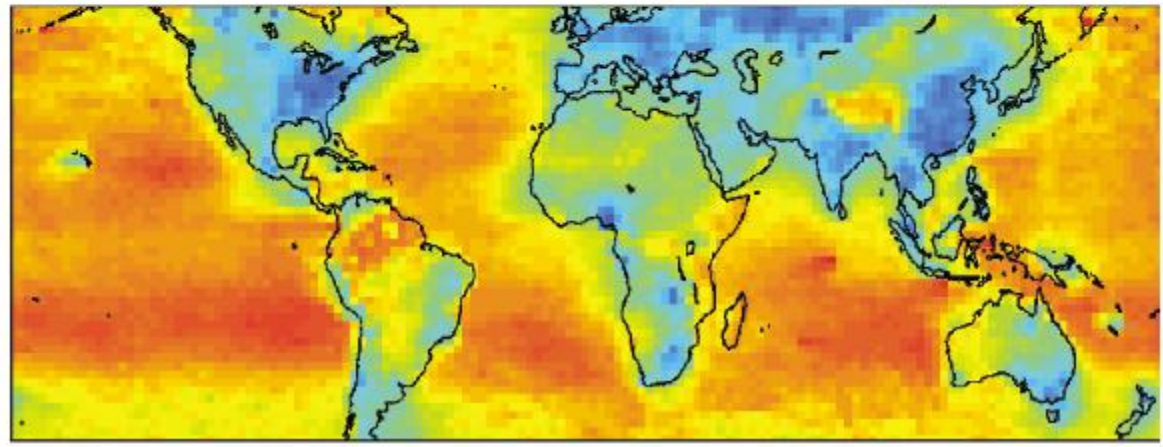
IV. Air mass factor (AMF) for conversion of tropospheric slant columns to tropospheric vertical columns

2005 mean AMF for SCIAMACHY tropospheric NO₂



$$VC = \frac{SC}{AMF}$$

2006 mean AMF for SCIAMACHY tropospheric SO₂



[Lee, C. et al., 2009]



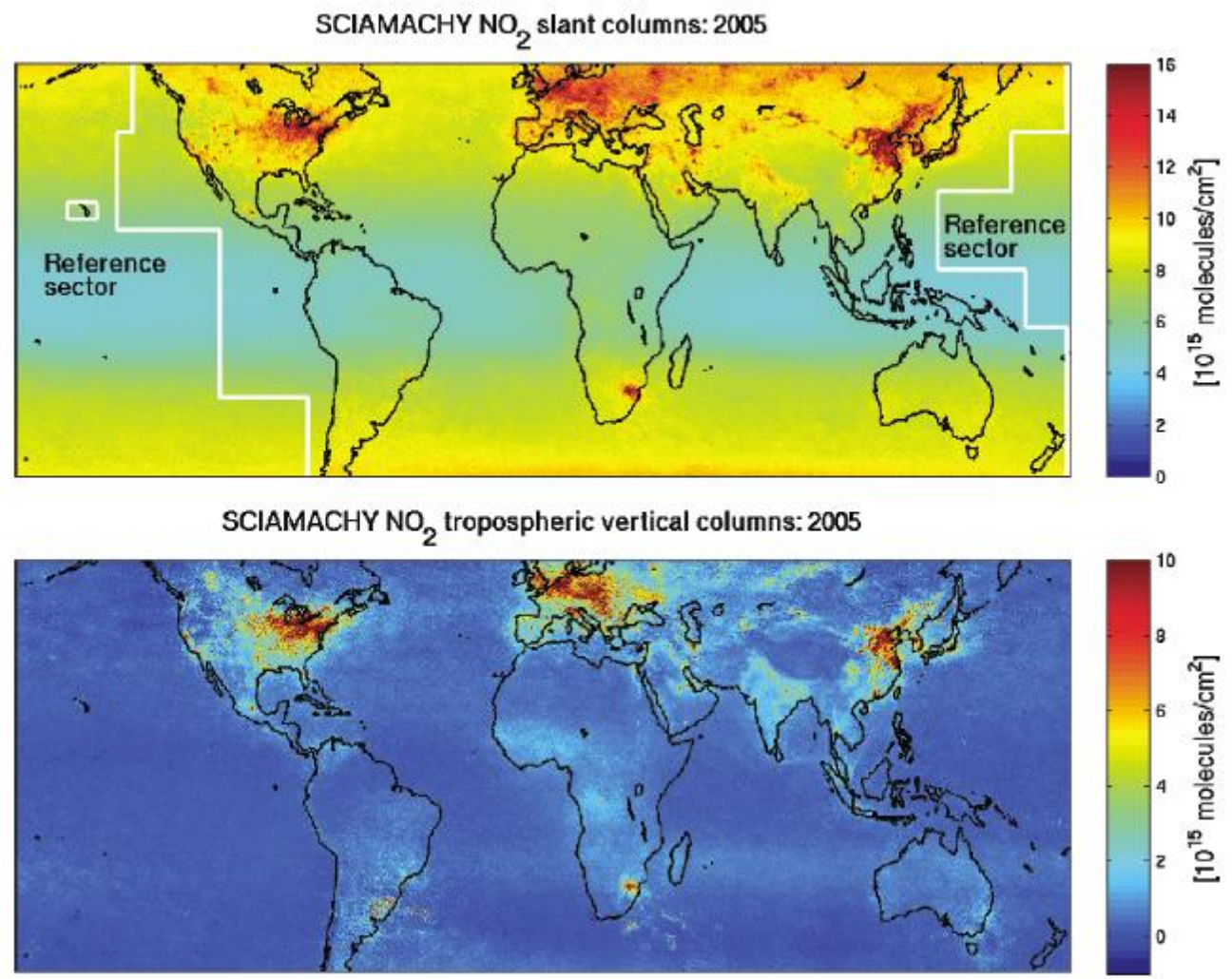
IV. Specifications of NO₂ and SO₂ retrievals for SCIAMACHY data

Species	Wavelength range (nm)	Polynomial order	Cross-sections included in the fit
NO ₂	426–450	3	NO ₂ at 243 K (Vandaele et al. 2002) O ₃ at 223 and 243 K (Bogumil et al. 2003) H ₂ O at 296 K (Rothman et al. 2005) O ₂ -O ₂ at 296 K (Greenblatt et al. 1990) Ring (Chance and Spurr 1997)
SO ₂	315–327	4	SO ₂ at 295 K (Vandaele et al. 1994) O ₃ at 223 and 243 K (Bogumil et al. 2003) Ring (Vountas et al. 1998) Undersampling Polarization dependency

[Lee, C. et al., 2009]



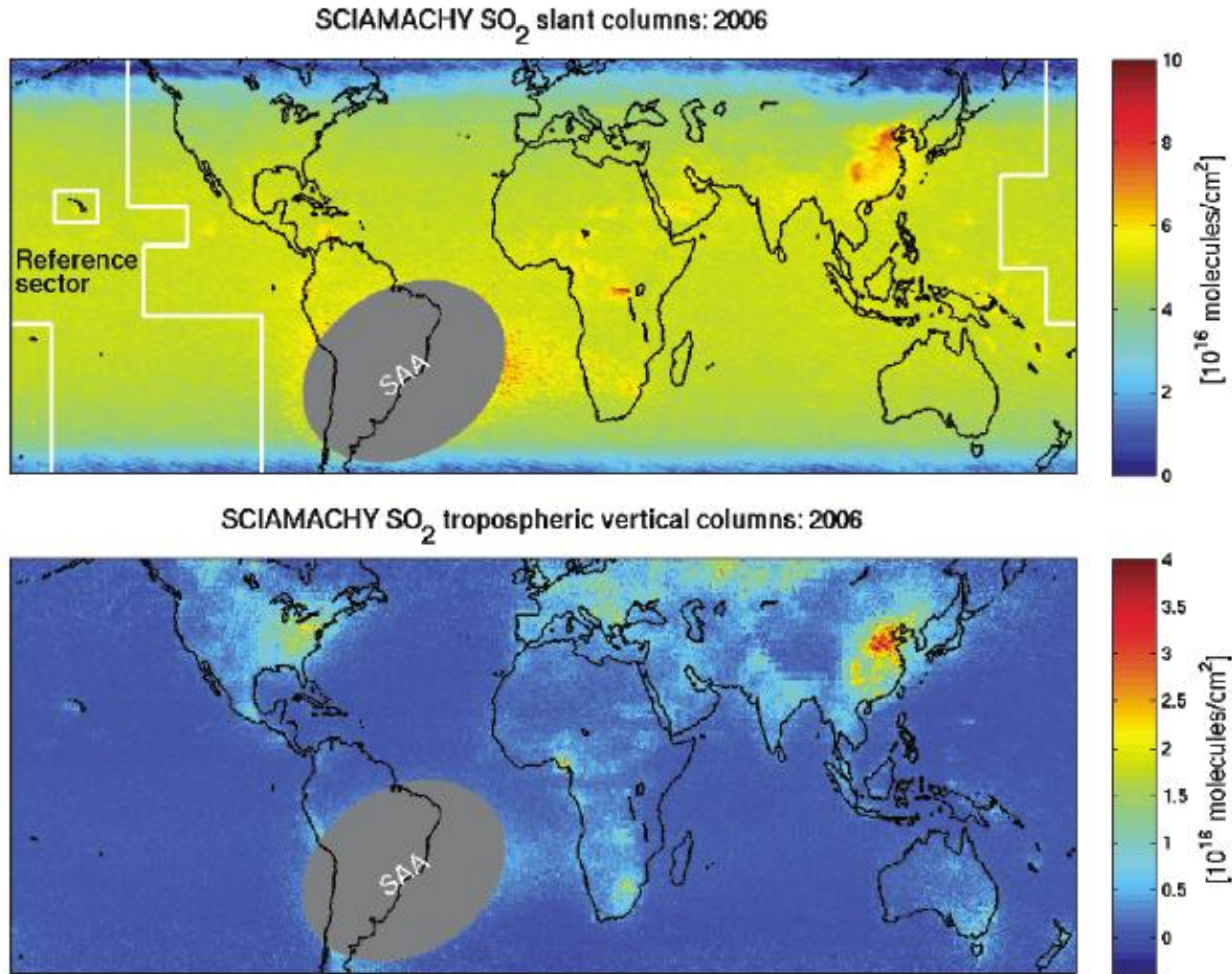
IV. NO₂ slant column and tropospheric vertical columns retrieved from SCIAMACHY



[Lee, C. et al., 2009]



IV. SO₂ slant column and tropospheric vertical columns retrieved from SCIAMACHY.



[Lee, C. et al., 2009]

IV. Summary

- Global mapping of trace gases from space provides critical information for constraining their emissions and improving our understanding of tropospheric chemistry.
- The retrievals of NO₂ and SO₂ in the troposphere and the AMF calculation to convert slant columns to vertical columns were presented.
- Satellite remote sensing has been successfully applied to measurements of trace gases in the troposphere.
- Satellite measurements of trace gases in the atmosphere are a crucial step forward for real time monitoring of air quality and forecasting such as hazard warning on a global scale.

