

Answers to referees' comments:

« Transboundary ozone pollution across East Asia: daily evolution and photochemical production analysed by IASI+GOME2 multispectral satellite observations and models”
by Juan Cuesta et al.

Dear referees,

We would like to thank you very much for your remarks that have improved the clarity of the paper. In the Revised Manuscript, called RM hereafter, we have addressed in detail each of your comments by adding new explanations in the manuscript and some minor modifications in the figures. All the recommendations of the reviewers have been followed and all clarifications were provided. Please, find below the detailed answers and how they are introduced in the manuscript.

Anonymous Referee #1

Received and published: 23 November 2017

General Comment

The authors examined a temporal evolution of ozone in a transboundary pollution event occurred in early May 2009 over East Asian countries by using multiple satellite observations and chemical transport models. The use of multispectral satellite data of IASI and GOME2 provides LMT ozone concentration, which cannot be obtained by single-band retrievals. They clearly showed how well the IASI+GOME2 approach retrieve the ozone concentration in LMT and applied it to describe an outbreak of transboundary ozone pollution event in East Asia. The large-scale observation of ozone near the surface (LMT) from the satellite with this approach is apparently a powerful tool for the air quality researches over the globe. This paper is well within the scope of the journal, however, I noticed several issues in this paper, which cannot be passed over to be published. I suggested that the authors should consider the following comments: one major and several specific comments.

Major Comment:

My biggest concern is the arbitrariness in the use of model results. The authors used the simulation with two different chemistry transport models to explain the daily evolution of ozone pollution across East Asia. I suppose that both of the two models can simulate the same chemical quantities such as the three-dimensional concentration of O₃, CO, NO₂. However, the authors did not fully utilize the results from both models, but they only used the result from one of the models for one quantity in most cases. I don't think it is fair to arbitrary pick up only the better and propitious result from one of the models for their interpretation. I strongly suggest the authors to evenly use the results from both models for each chemical quantity. To put my point differently, the author should clearly state the different roles of the different models that they supposed at the very first part of the paper, so that the reader may not feel arbitrary use of the models.

Clarified and completed.

This aspect is fully clarified in the RM and Figure 14 is completed for consistency. Three-dimensional fields of O₃, CO and NO₂ are available from both WRF-Chem and CHASER

models. However, using both models in all cases would largely increase the number of figures, without adding much relevant information for the paper. Therefore, the criteria to choice between the models used in the figures of the paper are the following: i) WRF-Chem is used to describe the structure of the plumes of LMT ozone and carbon monoxide with finer spatial resolution (Figures 4c-f, 6a-c, 10d, 11d and 12d), ii) CHASER forecasts with and without stratospheric ozone allows a distinction between tropospheric ozone originating from the stratosphere and the troposphere (Figs. 6e-f, 10f, 11f, 12f, 14e-f), iii) both WRF-Chem and CHASER analyses are used for showing the temporal lagrangian evolution of O₃, CO and NO₂ for the polluted air masses (Figure 13b-c and 14a-d,j) and iv) in the comparison of IASI+GOME2 and in situ data (Figure 2), CHASER analyses indicate the corresponding vertical gradient of ozone between the surface and 2 km of altitude with presumably good absolute accuracy provided by assimilation of various observations. For consistency in Fig. 14, we have added the time series of O₃, CO and NO₂ concentrations (Figs. 14c, d and j respectively) from CHASER analyses.

This clarification is added in the RM as follows (lines 28-32, page 12, lines 1-4, page 13):
“In the figures of the paper, we show one of the models or both of them according to the following criteria: i) WRF-Chem describes the structure of plumes of LMT O₃ and CO with finer spatial resolution (section 3), ii) CHASER forecasts with and without stratospheric ozone distinguish tropospheric ozone formed at the troposphere from that originating from the stratosphere (sections 3 and 4), iii) both WRF-Chem and CHASER are used for showing the temporal Lagrangian evolution of O₃, CO and NO₂ for polluted air masses (section 4) and iv) CHASER analyses indicate the vertical gradients of ozone between the surface and 2 km of altitude with presumably good absolute accuracy provided by assimilation of various observations (section 2).”

Specific Comments:

- Abstract: The definition of lowermost tropospheric ozone should be provided.

Done.

The definition is provided as (line 16 of page 1): “ ... lowermost tropospheric ozone (located below 3 km of altitude)”

- P3 L18-19: A brief explanation why single-band retrieval cannot provide the information in PBL is better here, not mandatory though.

Clarified.

The following brief clarification is provided in the RM (lines 19-23 of page 3): *“Standard single-band ozone retrievals cannot provide quantitative information at the planetary boundary layer (PBL), but at lowest at the lower troposphere (LT, i.e. below 6 km of altitude). Sensitivity to ozone for these retrievals essentially peaks at the free troposphere above the PBL, according to the available information on near-surface ozone.”*

-P6 L18-19: I could not find IASI+GOME2 data provided in both URL.

Corrected. The URL has been updated.

You can now find a description of the IASI+GOME2 data and the way to obtain it at <http://cds-espri.ipsl.fr> The general portal of the data centre <https://www.aeris-data.fr>

redirects the user to <http://cds-espri.ipsl.fr> . It is important to mention both URLs.

Data availability is better described in the RM as (line 32, page 6 and lines 1-2, page 7): *“global scale IASI+GOME2 retrievals are routinely produced by the French data centre AERIS and they are publicly available (see <https://www.aeris-data.fr> and <http://cds-espri.ipsl.fr>).”*

- P6 L28: Should more clearly describe the criteria of special coincidence. Is one degree lat/lon criteria between the location of sonde station and the center point of the satellite visual field?

Clarified.

Yes, it is one-degree latitude/longitude between the location of launching station of the sonde and the centre point of the satellite pixel.

This is clarified in the RM as (lines 11-13 page 7): *“Coincidence criteria are spatial colocalization of one-degree latitude/longitude between the locations of the launching stations of the sondes and the centre points of satellite pixels”.*

- Table 1: This table is not referred in the manuscript. If it is not necessary for the paper, it's better to remove it.

Corrected.

Table 1 is useful for the paper as it allows the reader to quickly and easily read the results of the comparison between ozonesondes and the satellite retrieval.

The reference to Table 1 has been added in the RM as (lines 4-5 page 7): *“An assessment of the quality of IASI+GOME2 for retrieving LMT ozone is presented in Figure 1 and summarized in Table 1”.*

- Figure 1: Is the symbol “ 1σ ” in legend widely used? I think at least a brief explanation is necessary in the caption.

Clarified.

For clarity in the RM, the symbol has been changed to $\sigma_{\text{sonde}}/\sigma_{\text{sat}}$ and the captions of Figure 1 and 2 explain it as: *“The symbol $\sigma_{\text{sonde}}/\sigma_{\text{sat}}$ is the ratio between the standard deviations of the sonde data and the satellite retrievals.”*

- P7 L19: Why the sonde stations in Asia outside Japan, such as Hong Kong or Hanoi, were ignored? They have data for the year 2009 and 2010.

Clarified.

Sonde data from Hong Kong and Hanoi stations is included in the Worldwide comparison (Fig. 1a and Table 1). The region of Asia analysed in the paper is 25-45°N 110-150°E. Hong Kong and Hanoi are located south of 25°N and therefore they were not included in the East Asian analysis.

This aspect is clarified in the RM as (line 32, page 7 and line 1 page 8): “As this paper focuses on East Asia (particularly at 25-45°N 110-150°E), we also present the comparison for all sondes available over this region ...”

- P7 L28: EANET and GAW are different NW, so you should refer to them separately here. Also you should provide the source URL for GAW database.

Corrected.

Both networks are referred in RM separately as follows (lines 11-14 page 8): “from 9 stations of the EANET (Acid Deposition Monitoring Network in East Asia, <http://www.eanet.asia>) network over East Asia, one station from the GAW (Global Atmosphere Watch, <http://www.wmo.int>) network”

- P8 L10: Is “vertical difference” more appropriate to this quantity than “vertical gradient” here. If you prefer to use gradient, it is natural to me that the unit is ppb/km.

Corrected.

We use in the RM the unit ppb/km for the vertical gradient, as follows (lines 26-27 page 8): “vertical gradients $\Delta O_3^{surf-2km}$ between the surface and 2 km of altitude lower than ± 10 ppb/km”

- P8 L18-19: The author can discuss about the following paper here, since the paper also tried to retrieve the lower tropospheric ozone from OMI data.

Hayashida S., Liu X., Ono A., Yang K., Chance K., 2015. Atmospheric Chemistry and Physics 15, 9865-9881.

Done.

We have mentioned the lower tropospheric ozone retrieval from OMI in this paragraph of the RM as follows (lines 22-25 page 9): “This IASI retrieval is often used to analyse ozone enhancements at the lower troposphere over Europe (e.g. Eremenko et al., 2008) and East Asia (e.g. Dufour et al., 2015), as also done with retrievals from OMI measurements also over East Asia (Hayashida et al., 2015).”

- Figure 3: Fig3a and b shows the LMT O3 in unit of ppb. Is this an average concentration of O3 in the lowermost 3 km altitude? If so, you should clearly state it somewhere in the manuscript (and the caption of this figure).

Clarified.

In the text of the RM and the caption of Fig. 2, we clearly state is as (line 32 page 8 and lines 1-3 and page 8): “Ozone concentrations at the LMT are provided as volume mixing ratios in ppb (parts per billion), calculated as the ratio of LMT partial columns (in molecules per cm²) of ozone and air (in Figures 2-14 and also used for CO and other partial columns).”

- P12 L6-10: The version of each EI should be described.

Done.

In the RM, we have added the version of each Emission inventory (lines 10-13 page 13): “Surface emissions over China and North and South Korea are taken from REAS (Regional Emission Inventory in Asia; Ohara et al. 2007) version 1.11, and over Russia from EDGAR (Emission Database for Global Atmospheric Research; Olivier et al. 1996) version 3.2.”

- P12 L12: Is this an appropriate reference for CHASER model?

Corrected.

For the CHASER model, the RM use the following references (lines 16-17 page 12): “Sudo et al., 2002; Sudo and Akimoto, 2007; Sekiya and Sudo, 2014”

Sudo, K., Takahashi, M., Kurokawa, J., and Akimoto, H.: CHASER: A global chemical model of the troposphere 1. Model description, J. Geophys. Res., 107, 4339, <https://doi.org/10.1029/2001JD001113>, 2002.

Sudo, K. and Akimoto, H.: Global source attribution of tropospheric ozone: Long-range transport from various source regions, J. Geophys. Res., 112, D12302, <https://doi.org/10.1029/2006JD007992>, 2007.

Sekiya, T. and Sudo, K.: Roles of transport and chemistry processes in global ozone change on interannual and multidecadal time scales, J. Geophys. Res., 119, 4903–4921, <https://doi.org/10.1002/2013JD020838>, 2014.

- P12 L23-25: Is this reduction of bias for column density? How about the reduction of bias for the surface concentration by assimilation?

Clarified.

The reductions in biases by data assimilation are enounced in column for NO₂ and concentrations for CO in the lower troposphere and O₃ in the middle and upper troposphere.

This aspect is fully clarified in the RM as (lines 25-31 page 13): “It reduces biases for tropospheric NO₂ columns by 40–85 %, for lower tropospheric CO concentrations in the Northern Hemisphere by 40–90 % and for O₃ in the middle and upper troposphere by 30–40%. Data assimilation also mostly removed the model’s negative bias in surface CO concentrations in the northern hemisphere. The error reduction for O₃ was generally smaller in the lower troposphere than in the middle and upper troposphere because of the reduced sensitivity of the assimilated TES retrievals to lower-tropospheric ozone.”

- P13 L2-4: Did you do assimilation for the sensitivity analysis too? If so, can we assume the influence of assimilation process is the same for both full and sensitivity simulation? If the impact of assimilation is different in two simulations, the difference of ozone concentration between full and sensitivity simulation cannot be regarded as a pure stratospheric contribution.

Clarified.

Assimilation is not performed in the simulations used in the sensitivity analysis. The difference is calculated between two simulations run in forecast mode. The difference

between the 2 simulations is therefore consistent for estimating tropospheric and stratospheric contributions of ozone.

The RM clarifies this issue as (lines 21-22 page 14): *“For consistency, no data assimilation is performed in either of the two simulations of this sensitivity analysis.”*

- P13 L8: Is this reanalysis (ERA-INT) used to drive CHASER?

Clarified.

CHASER is driven by AGCM meteorological fields nudged toward NCEP-DOE/AMIP-II reanalyses at every time step of the AGCM to reproduce past meteorological fields.

This aspect is indicated in the RM as (lines 9-12 page 14): *“The AGCM fields are nudged toward the National Centers for Environmental Prediction/Department of Energy Atmospheric Model Intercomparison Project II (NCEP-DOE/AMIP-II) reanalysis (Kanamitsu et al., 2002) at every time step of the AGCM to reproduce past meteorological fields.”*

- P14 L16: You should describe clearly how to set the magenta rectangle in the figures. Are there any objective criteria to draw the four sides of the rectangle? There is no description of the rectangle in the caption of Figure 4 and 6.

Clarified.

Magenta and red rectangles in Figures 4-12 are rectangular zones containing all valid satellite pixels (gridded $1^\circ \times 1^\circ$) used to describe the daily evolution of polluted air masses originating from the North China Plain and travelling to the Pacific. These satellite pixels are those co-located with at least 5 % of the polluted air parcels trajectories simulated by HYSPLIT dispersion model.

In the RM, this is clarified as follows (lines 16-19 page 16): *“In Figures 4-12, satellite pixels used to describe the evolution of these polluted air masses are depicted by magenta and red rectangles. These boxes contain valid satellite pixels co-located with at least 5 % of the polluted air parcels trajectories simulated by Hysplit.”*

- P16 L9-10: Where is “this location” here?

Clarified.

This statement on Fig. 6b refers to the region $40-45^\circ\text{N}$ $122-128^\circ\text{E}$ (previously commented for Fig. 5b).

The RM indicates is as (line 31 page 17): *“At this location ($40-45^\circ\text{N}$ $122-128^\circ\text{E}$)”*

- P16 L16-17: I cannot agree with here, for me, Fig 6b is not so good agreement with Fig 5a.

Corrected.

Indeed, differences in LMT ozone simulated by WRF-Chem and retrieved by IASI+GOME2 are seen north of 38°N ($110-120^\circ\text{E}$) with higher values for satellite

retrievals. However, both WRF-Chem and IASI+GOME2 show relatively high LMT ozone concentrations over the NCP.

This is corrected in the RM as (lines 6-8 page 18): *“As previously mentioned, IASI+GOME2 also retrieves high LMT ozone concentrations over the NCP (around 35°N 115°E, Fig. 5a), but also north of it (differing from simulations at the LMT).”*

- P16 L23-25: Which figure does this sentence mention to?

Clarified and Corrected.

This sentence refers to the general method used for all figures (Figure 4-12) where the polluted air masses are tracked. In this paragraph, it is indeed misleading. As it is also redundant with section 2.4, the referred sentence is erased from the RM.

- P17 L26: Should clarify how to initiate the Hysplit calculation for two pollution plumes.

Clarified.

In our analysis, we initialize Hysplit at two starting locations separated by a relatively small distance, which suggests two different trajectories for the polluted air masses. The trajectory of the southern plume (magenta in Fig. 9) is obtained with the mean location of the air parcels of the previous day and that of the northern plume (red in Fig. 9) by initializing Hysplit 2 degrees northeast from that position. The existence of two different LMT ozone plumes is clearly evidenced by IASI+GOME2 observations in the following days (Fig. 12) and is also confirmed by trajectories in backward mode initiated at the location of these plumes.

In the RM, we clarify this aspect as (lines 21-27 page 19): *“The trajectories of the southern and northern pollution plumes (respectively magenta and red in Fig. 9) are obtained by initializing Hysplit respectively at the mean arrival location of the trajectories from the previous day and 2 degrees northeast from that position. The common geographical origin of the two plumes (before 6 May) is confirmed by Hysplit trajectories in backward mode initiated at the location of the two major ozone plumes clearly observed by IASI+GOME2 (e.g. south and north of Japan two days after in Fig. 12a).”*

- P17 L27: Typo? (Fig. 9a)

Corrected.

Both Fig.9a and 9c show the two depicted pollution plumes, respectively with boxes and the actual location of the Hysplit air parcels. In the RM, we indicate it as (line 17 page 19): *“Fig. 9a,c”*.

- P18 L17: I cannot see the enhanced NO₂ at the area pointed out here. Is this sentence correct?

Clarified.

The enhancement of NO₂ with respect to the background at the region 43-45°N 126-132°E is only moderate (up to $6 \cdot 10^{15}$ mol/cm²). Therefore, the sentence is changed in

the RM as (lines 14-15 page 20): *“as suggested by moderately enhanced NO₂ concentrations at 43-45°N 126-132°E up to 6 10¹⁵ mol/cm² in Fig. 10c”*.

- Figure 10e: The altitude of PV contour (300 hPa) should be described in the caption. - P19 L21 & L28: Typo? 11e and 12e ?

Corrected.

In the RM, we have added *“at 300 hPa”* in the caption of Figure 10e and corrected the typos on the figures. For CHASER, we indicate Fig.11f and Fig.12f and for PV Fig.11e and Fig.12e.

- P20 L1: 11f and 12f?

Corrected.

The RM indicates Fig. 11f and 12f.

- P21 L5: Fig 13 should be Fig 13a.

Corrected.

The RM indicates Fig. 13a (line 5 page 23).

- P22 L4: NO titration

Corrected.

The RM indicates (line 4 page 24) *“nitrogen monoxide (NO) titration”*.

- P22 L6: Only horizontal dilution is important?

Clarified.

Since the reduction in CO concentrations are remarked during only 3 days and the typical lifetime of CO is in the order of a few months, the only remaining factor for reducing CO at the LMT is atmospheric dilution (horizontal and/or vertical).

Since it is not only horizontal dilution, the statement in the RM has been changed to (lines 8-9 page 24): *“... linked to atmospheric dilution (horizontal and/or vertical). Sinks of CO are not expected to be significant during a period of 3 days.”*

- P22 L12: I don't think the absolute values of the ratio dO₃/dCO are consistent with each other from satellites observations and models. The models show apparent lower ratio than the observation. Please clarify what aspect is consistent each other.

Agreed and corrected.

We agree with your remark that in absolute values, the ratio $\Delta O_3/\Delta CO$ derived from the satellite measurements is higher than that from models, particularly at the beginning of the period (before 6 May).

This is modified in the RM (lines 13-17 page 24): *“In absolute values, the ratio $\Delta O_3/\Delta CO$ derived from the satellite measurements is higher than that from models. At the beginning of the event (3-5 May), satellite estimates of the ratio are 0.1 to 0.15 higher than those*

from satellite. After 6 May, satellite and WRF-Chem ratios are closer (with differences between 0.05 and 0.1)."

- P22 L14: Typo? "those from models"?

Corrected.

Indeed, it is "*those from models*" as corrected in the RM (line 16 page 24).

- P22 L21: It is not easy to understand the meaning and implication of "degrees of freedom" for the readers outside the satellite data analysis. It's better to briefly explain them here.

Clarified.

The following description of the "degrees of freedom" of the satellite retrievals is provided in the RM (line 10 page 3): "*degrees of freedom (i.e. the number of independent pieces of information of the retrieved profile)*" and (lines 25-28 page 24): "*This is described in terms of the degrees of freedom (i.e. the number of independent pieces of information) and the altitude of maximum sensitivity of the retrieved atmospheric columns, which respectively quantify the amount of information provided by the satellite retrieval and the altitude it comes from.*"

- P22 L 25-26: Which figure does this sentence mention to? - P22 L29: What is "3-9" here?

Clarified.

This sentence comes from Figure 14h of the RM. The word "May" is missing after "3-9". These corrections are included in the RM (line 5 page 25).

- P23 L1-2: I don't think the stratospheric contribution remains constant in anyway. It fluctuates a lot during the period.

Agreed and corrected.

We agree that the stratospheric contribution at the upper troposphere fluctuates significantly. This mistake was corrected in the RM as (lines 11-13 page 25): "*On the other hand, the ozone contribution of stratospheric downward transport at the upper troposphere (from 6 to 12 km asl) fluctuates significantly during the whole event (Fig. 14f).*"

- P23 L7-10: It is not easy to see consistency in the ozone partial column between satellite observation and model (CHASER) simulation. It's better to use more words to describe which aspect do you think is consistent to each other.

Clarified.

A fair consistency between IASI+GOME2 and CHASER is remarked in the average concentrations of ozone at the LMT and the upper troposphere.

We clarify it in the RM as (lines 15-18 page 25): "*we remark that similar concentrations of ozone at the LMT and the upper troposphere are retrieved by IASI+GOME2 and simulated by CHASER (adding contributions from the Troposphere and Stratosphere in) averaged over the whole event (differences of 13 ppb at most).*"

- P23 L16-17: How does the concentration change in NO₂ on 7 May simulated by WRF-Chem? Is it similar to what simulated in CHASER?

Clarified.

Yes, as shown in the new figure 14j of the RM, NO₂ concentrations simulated by both WRF-Chem and CHASER on 7 May show very similar relative temporal evolutions, with a clear enhancement with respect to the previous day. The similarity in the evolution of NO₂ simulated by both model is kept over whole period (3-9 May) in relative terms, but LMT NO₂ concentrations in absolute values are a factor 3 higher for WRF-Chem than for CHASER.

In the RM, this is written as (lines 9-11 page 26): “Both WRF-Chem and CHASER simulations suggest a relatively higher availability of NO₂ at the LMT for the northern pollution filament” and (lines 4-5 page 26) “The latter might be linked to low availability of NO₂ at the LMT in CHASER simulations (a factor 3 lower than for WRF-Chem, Fig. 14j)”

- P23 L23: Typo. red curve -> blue curve

Agreed and corrected.

- P24 L2-5: The authors referred to relatively high NO₂ concentration in CHASER as a cause of greater growth of dO₃/dCO ratio in the northern plume than the southern plume from 7 to 9 May. However, the ratio in CHASER did not show such a growth both in the northern and southern plumes (Figure 13c). So it is not appropriate to refer to the change in NO₂ in CHASER as a cause of the observed change in the ratio.

Agreed and clarified.

We agree that only CHASER simulations are not sufficient evidence to analyse the observed growth of dO₃/dCO. However, WRF-Chem shows the same relative increase as CHASER in NO₂ concentrations on 7-9 May, and higher by a factor 3. Therefore, we mention the enhancement of LMT NO₂ for both WRF-Chem and CHASER simulations (3 times higher for WRF-Chem) that suggest a higher availability of NO₂ to explain the observed change in the ratio.

In the RM, this is written as (lines 9-13 page 26): “Both WRF-Chem and CHASER simulations suggest a relatively higher availability of NO₂ at the LMT (although 3 times higher for WRF-Chem) for the northern pollution filament (dotted curves with respectively blue ovals and green stars in Fig. 14j) as for the southern plume (light blue and light green in Fig. 14j).”

- P24 L17-19: Can you estimate the impact of this effect on the dO₃/dCO ratio quantitatively? Excluding under- or over-estimation of dO₃/dCO ratio due to the change in the sensitivity of satellite retrievals is quite important to make this approach useable for O₃ production estimation during air mass transport.

Done and clarified.

We have done a sensitivity analysis of the errors in dO_3/dCO with respect to changes in sensitivity of the satellite retrievals. For this, we have used model outputs of typical vertical profiles of O_3 and CO for a pollution plume and smoothed them with typical averaging kernels of the satellite retrievals. We have taken into account that heights of maximum sensitivity usually change concomitantly for both O_3 and CO retrievals, as in both cases it depends on thermal contrast between the surface and the air. The results show that under and overestimations of dO_3/dCO remain below $\pm 11\%$ for changes of 1 and 3 km in the heights of maximum sensitivity for respectively O_3 and CO . This uncertainty is significantly lower than changes observed for dO_3/dCO from satellite retrievals (up to 80% during *the whole event*). Therefore, the proposed approach is valid for estimating O_3 production during air mass transport.

These results and clarifications are provided in the RM as (lines 27-32 page 26 and lines 1-5 page 27): *“According to sensitivity analyses, these uncertainties induce under or overestimations for $\Delta O_3/\Delta CO$ that remain below $\pm 11\%$ for changes of 1 and 3 km in the heights of maximum sensitivity for respectively O_3 and CO retrievals. These estimations are obtained using typical vertical profiles of O_3 and CO for a pollution plume (from WRF-Chem) smoothed with averaging kernels of the satellite retrievals and taking into account the concomitant change in the heights of maximum sensitivity for O_3 and CO retrievals, as in both cases they depend on the difference between surface and air temperatures. These uncertainties are significantly lower than changes observed for $\Delta O_3/\Delta CO$ from satellite retrievals (up to 84% during the whole event). Therefore, conclusions drawn on the occurrence and quantification of photochemical ozone production in this period are not significantly affected by changes in satellite retrievals sensitivities.”*

Anonymous Referee #2

Received and published: 3 March 2018

This paper elaborates a comprehensive study of transboundary ozone pollution across East Asia via employing [1] the multiple-spectral IASI/GOME2 ozone profile product that provides the quantitative estimates of ozone concentration in the LMT; and [2] the combined modeling tools consisting of CHASER (global scale) and WRF-CHEM (regional scale) models. This study provides multi-species, multi-scale picture of pollutions across East Asia, helping in distinguishing between local and non-local drivers of pollution in LMT. The subject of the paper is appropriate to ACP. Below are a few comments concerning clarifications/extensions for consideration in the final publication.

This work uses the IASI carbon monoxide (CO) profile data to estimate the CO concentration in lower troposphere (LT), then use IASI LT CO and joint IASI+GOME2 LMT O3 as daily pictures for facilitate the study of daily evolution of pollution across East Asia.

1) The authors should describe how well the IASI LT CO data could represent the CO variability in the LMT.

Clarified.

The approach developed by ULB/LATMOS retrieves lower tropospheric CO (below 6 km of altitude) from IASI with 0.83 degrees of freedom and a height of maximum sensitivity around 4.7 km of altitude asl, in average over the region and period studied in the paper. This CO product is sensitive at the LMT (up to 3 km of altitude asl) with 0.51 degrees of freedom in average. In our paper, we use LT CO retrievals as they are already validated against MOZAIC in situ measurements (De Wachter et al., 2012). A dedicated validation of other CO partial columns against independent measurements is beyond the scope of the present paper, as it is mainly focused on ozone pollution.

In the RM, this is written as (lines 2-5 page 11): *“LT partial columns are retrieved with 0.83 degrees of freedom (DOF, i.e. number of independent pieces of information in the retrieved profile) in average over the region and period studied in the paper. This product provides significant information on CO variability below 3 km asl, as DOF at the LMT are 0.51 on average.”*

2) Drs. Miyazaki and Sekiya have developed a high-resolution CHASER simulation tool (version 4.0) with a finest spatial resolution of 0.56 degrees (Sekiya et al., 2017) – significantly higher than that of CHASER and WRF-Chem models used in this study. The performance of CHASER v4 has been validated using reference data sets from satellite missions and aircraft flight campaigns. The authors should include this reference in this paper and provides some discussions.

Sekiya T., Miyazaki K., Ogochi K., Sudo K., and Takigawa M., Global high-resolution simulations of tropospheric nitrogen dioxide using CHASER V4.0, Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2017-203>, in review, 2017.

Done.

Indeed, Sekiya et al. (2018) developed a high-resolution version of the CHASER model at 0.56° resolution and demonstrated the improved model performance over areas with strong local sources by increased the horizontal model resolution from 2.8° to 0.56°. Nevertheless, the 2.8° resolution model is capable to simulate synoptic ozone patterns.

The RM provides this information as follows (lines 2-6 page 14): “Sekiya et al. (2018) have recently developed a high-resolution version of CHASER with 0.56° horizontal resolution and demonstrated improved performances over areas with strong local sources with respect to the 2.8° resolution version. Nevertheless, the CHASER model with 2.8° resolution is capable of properly simulating synoptic ozone patterns.”

3) Page 4, Line 5-6: There is a multiple spectral retrieval algorithm developed for CO profile retrievals (Fu et al. 2016). They demonstrated the feasibility of combining the measurements from Sentinel-5 precursor (S5P) TROPOMI (near infrared) and Suomi-NPP (SNPP) CrIS (thermal Infrared) sensors to extend Terra MOPITT both TIR alone and multiple spectral CO profile products capable of quantifying the first 2-3 km CO amounts, as well as improving spatial coverage and resolution in comparison to Terra-MOPITT. The authors could add some discussions nearby the end of first/beginning of second paragraphs of page 4, e.g, “The Sentinel-5 precursor (S5P) and Suomi NPP (SNPP) has successfully formed a new satellite constellation, leading to a unique opportunity to quantify the amounts of carbon monoxide in the LMT over global scale via combining the satellite measurements from SNPP CrIS (TIR) and S5P TROPOMI (NIR) instruments. Fu et al. (2016) presented the methodology and characteristics of joint CrIS/TROPOMI CO profile retrievals, demonstrating the feasibility of extending the decadal record of Terra-MOPITT CO products (Worden et al., 2010 and 2013).”

Fu D., Bowman K.W., Worden H., Natraj V., Yu S., Worden J.R., Veefkind P., Aben I., Landgraf J., Strow L., Han Y., High resolution tropospheric carbon monoxide profiles retrieved from CrIS and TROPOMI, Atmos. Meas. Tech., 9, 2567-579, 2016.

Worden H.M., Deeter M.N., Edwards D.P., Gille J.C., Drummond J. R., and Nédélec, P. P., Observations of near-surface carbon monoxide from space using MOPITT multi-spectral retrievals, J. Geophys. Res., 115, D18314, doi:10.1029/2010JD014242, 2010.

Worden H.M., Deeter M.N., Frankenberg C., George M., Nichitiu F., Worden J., Aben I., Bowman K. W., Clerbaux C., Co-heur P.F., de Laat A.T.J., Detweiler R., Drummond J. R., Edwards D.P., Gille J. C., Hurtmans D., Luo M., Martínez-Alonso S., Massie S., Pfister G., and Warner J.X., Decadal record of satellite carbon monoxide observations, Atmos. Chem. Phys., 13, 837–850, doi:10.5194/acp-13-837-2013, 2013.

Done.

The 3 references have been added as well as the following paragraph in the RM (lines 9-20 page 4): “Multispectral synergisms are also implemented to retrieve other atmospheric species with enhanced near-surface sensitivity, as carbon monoxide (CO). This is done with measurements in the thermal and near infrared from the Measurements Of Pollution In The Troposphere (MOPITT) instrument onboard the Earth Observing System (EOS) Terra satellite (Worden et al., 2010). Recently, Sentinel-5 precursor (S5P) and Suomi National Polar orbiting Partnership (SNPP) have successfully formed a satellite constellation,

leading to a new opportunity to quantify the amounts of CO at the LMT over global scale by combining the satellite measurements in the thermal and near IR respectively from the instruments SNPP Cross-track Infrared Sounder (CrIS) and S5P TROPospheric Monitoring Instrument (TROPOMI). Fu et al. (2016) presented the methodology and characteristics of joint CrIS/TROPOMI CO profile retrievals, demonstrating the feasibility for extending the decadal record of MOPITT CO products (Worden et al., 2013)."

4) Page 38, Line 2, Figure 2 caption: IASI+GOME -> IASI+GOME2

Corrected.

1 **Transboundary ozone pollution across East Asia: daily**
2 **evolution and photochemical production analysed**
3 **by IASI+GOME2 multispectral satellite observations and**
4 **models**

5
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12

13 **Abstract**

14 We characterize a transboundary ozone pollution outbreak transported across East Asia in
15 early May 2009 using new multispectral satellite observations of lowermost tropospheric
16 ozone ([located below 3 km of altitude](#)) in synergy with other satellite data and models. Our
17 analysis is focused on the daily evolution of ozone pollution plumes initially formed over the
18 North China Plain (NCP) and their transport pathways over Northern China, Korea, Japan and
19 the surrounding seas. A main aspect of the study is an estimation of the contribution of
20 photochemical production of ozone along transport using the ratio of ozone to carbon
21 monoxide enhancements with respect to background levels derived from satellite data and
22 also from chemistry-transport models.

23 A key contribution of the analysis is the use of new satellite data offering unprecedented skills
24 to observe the horizontal distribution of lowermost tropospheric ozone over East Asia on
25 daily basis, with a multispectral approach called IASI+GOME2. These satellite observations
26 are in good agreement with ozonesondes, with low mean biases (3%), a precision of about
27 16%, a correlation coefficient of 0.85 and practically the same standard deviation for a
28 comparison based on 2 years of data from 46 launching stations distributed worldwide, during

1 all seasons. A similar agreement is also found over East Asia. Moreover, IASI+GOME2
2 offers a unique capacity for observing the evolution of near surface ozone during pollution
3 outbreaks (with 5 % bias and 0.69 correlation), according to a comparison with surface in situ
4 measurements during 2 major ozone events over several Japanese Islands. Single-band ozone
5 retrievals, as those from IASI in the thermal infrared, do not capture such variability.

6 Using IASI+GOME2, we put in evidence that i) ozone pollution plumes are transported by an
7 anticyclonic circulation around the Yellow Sea from the NCP to Northern China, Korea and
8 Japan, co-located with carbon monoxide plumes, ii) over Northern China the plume splits into
9 two pollution filaments with one mixing with freshly emitted pollutants and iii) ozone is
10 produced every day of the event accounting for an enhancement in concentration during
11 transport across East Asia of up to ~84 % with respect to that produced over NCP. This
12 estimation is done according to monotonically increasing values during 7 days of the ratio of
13 ozone to carbon monoxide enhancements within the transported pollution plumes from about
14 ~0.25 over the NCP to ~0.46 over the Pacific south of Japan.

15

16 **1 Introduction**

17 Air pollution is now the world's largest single environmental health risk, causing 7 million
18 premature deaths worldwide every year (Lelieveld et al., 2015; World Health Organisation -
19 WHO, 2016). About 4.3 million of these deaths are related to ambient air pollution, from
20 which 2.6 million deaths per year occur over Southeast and East Asia as result of exposure to
21 the world's largest air pollution-related burden (WHO, 2016). East Asia, and in particular
22 China, experienced rapid economic growth (up to a factor 40 of the gross domestic product
23 since the 80's) and extensive urbanization during the last decades. Accordingly,
24 anthropogenic pollutant emissions have largely increased, making China one of the largest
25 pollution source regions in the world (Lu et al., 2011, Wang et al., 2013). In the main Chinese
26 megacities, ambient concentrations of the most harmful pollutants, such as tropospheric ozone
27 (O_3) and particulate matter (PM), largely exceed the thresholds recommended by WHO (Chai
28 et al., 2014). Air pollution originating from East Asia is also a worldwide-shared concern. It
29 can be transported and undergo chemical transformations far beyond country boundaries
30 within a day and around the hemisphere within one or two weeks, having a significant impact
31 on the budget of tropospheric pollutants at the intercontinental scale (e.g. Lin et al., 2010). For

1 example, current trends of ozone concentrations at the surface over Japan show a significant
2 increase, despite strong local controls of pollution emissions in the last decades, and probably
3 related to transboundary transport (Akimoto et al., 2015). Similarly, transcontinental transport
4 of Asian pollution probably explains the absence of reduction in ozone background levels
5 over the United States or Europe, despite local efforts for reducing the emissions of its
6 precursors (e.g. Dentener et al., 2010, Verstraeten et al., 2015).

7 The dramatic damages caused by East Asian air pollution at regional and intercontinental
8 scales strongly request for thorough monitoring of pollutant emissions both near the sources
9 and downwind from these regions, where secondary pollutants are photo-chemically
10 produced. However, the record of surface network observations of air pollution over China is
11 very limited, being openly available only since 2013 (Wang et al., 2014). On the other hand,
12 forecasting East Asian air pollution with chemistry-transport models is hampered by two
13 factors: insufficient surface observations for validating the simulations (particularly over the
14 East China Sea) and the lack of precision of the emission inventories, which are unable to
15 reflect the rapid changes in Chinese economy and the complexity of their emissions (Wang et
16 al., 2015).

17 Satellite observations offer a great potential for filling the observational gap of air pollution
18 over East Asia and overcome the limited spatial coverage of ground-based measurements.
19 Nevertheless, measuring ozone pollution from space is a challenging issue. Standard single-
20 band ozone retrievals cannot provide quantitative information at the planetary boundary layer
21 (PBL), but at lowest at the lower troposphere (LT, i.e. below 6 km of altitude). Sensitivity to
22 ozone for these retrievals essentially peaks at the free troposphere above the PBL, according
23 to the available information on near-surface ozone. Spaceborne spectrometers operating in the
24 UV, like OMI (Ozone Monitoring Instrument, Levelt et al., 2006) and GOME-2 (Global
25 Ozone Monitoring Experiment-2, EUMETSAT, 2006), have been used to derive tropospheric
26 ozone observations with sensitivity around 5-6 km of altitude (e.g. Liu et al., 2010, Cai et al.,
27 2012). Thermal infrared (IR) space-borne instruments, like IASI (Infrared Atmospheric
28 Sounding Interferometer, Clerbaux et al., 2009) on-board the MetOp satellites, have shown
29 good performance for observing ozone in the lower troposphere, but with sensitivity peaking
30 at 3 km of altitude at lowest (e.g. Eremenko et al., 2008; Dufour et al., 2012). Recently, a new
31 multispectral approach called IASI+GOME2, combining IASI observations in the IR and
32 GOME-2 measurements in the UV, allowed the first spaceborne observation of the full

1 horizontal structure and concentration of ozone plumes located near 2 km of altitude, for a
2 moderate European pollution outbreak (Cuesta et al., 2013). This approach offers the unique
3 capacity to observe the horizontal distribution of ozone in the lowermost troposphere (LMT),
4 hereafter defined as the atmospheric layer between the surface and 3 km of altitude above sea
5 level (asl). Similarly, the multispectral combination of TES (Tropospheric Emission
6 Spectrometer, Worden et al, 2007) and OMI measurements, respectively in the IR and UV,
7 has also shown an enhancement of sensitivity below 700 hPa (Fu et al., 2013), but with very
8 limited horizontal coverage (pixels longitudinally spaced by about 2000 km on the same day).
9 Multispectral synergisms are also implemented to retrieve other atmospheric species with
10 enhanced near-surface sensitivity, as carbon monoxide (CO). This is done with measurements
11 in the thermal and near infrared from the Measurements Of Pollution In The Troposphere
12 (MOPITT) instrument onboard the Earth Observing System (EOS) Terra satellite (Worden et
13 al., 2010). Recently, Sentinel-5 precursor (S5P) and Suomi National Polar orbiting
14 Partnership (SNPP) have successfully formed a satellite constellation, leading to a new
15 opportunity to quantify the amounts of CO at the LMT over global scale by combining the
16 satellite measurements in the thermal and near IR respectively from the instruments SNPP
17 Cross-track Infrared Sounder (CrIS) and S5P TROPOspheric Monitoring Instrument
18 (TROPOMI). Fu et al. (2016) presented the methodology and characteristics of joint
19 CrIS/TROPOMI CO profile retrievals, demonstrating the feasibility for extending the decadal
20 record of MOPITT CO products (Worden et al., 2013).

21 Simultaneously monitoring several air pollutants may offer useful insights on the origin and
22 evolution of ozone pollution. For example, high concentrations of both ozone and carbon
23 monoxide suggest an anthropogenic origin of the air masses, as CO is a primary product of
24 traffic and industrial emissions and is formed by oxidation of anthropogenic hydrocarbons. Its
25 lifetime is about 2 months (e.g. Logan et al., 1981). Tropospheric ozone-enriched airmasses
26 with background concentrations of CO and low water vapour levels are probably related to
27 downward transport from the stratosphere and the Upper-Troposphere/Lower-Stratosphere
28 (UTLS) region. The ratio between the enhancements of O₃ and CO with respect to the
29 background levels allows examining the production of ozone from combustion by-products
30 (nitrogen oxides - NO_x, hydrocarbons and CO) by photochemical processing of air parcels
31 during a few days to a week (e.g. Parrish et al., 1993, Chin et al., 1994, Mauzerall et al.,
32 2000). This approach may however underestimate ozone production along transport since CO
33 may not only be directly emitted but also produced by oxidation of hydrocarbons (Chin et al.,

1 1994; Gao et al., 2005). This ratio has been mainly estimated using in situ measurements at
2 several ground-based sites (Chin et al., 1994), from aircrafts (Price et al., 2004), model
3 simulations (Maurezall et al., 2000) and in a few cases with satellite data mainly sensitive at
4 the free troposphere (Zhang et al., 2006; Kim et al., 2013; Dufour et al., 2015). In addition,
5 high abundances of ozone precursors, such as nitrogen dioxide (NO_2) and volatile organic
6 compounds as formaldehyde (CH_2O), may be linked to higher photochemical production of
7 O_3 , depending on the regime of ozone atmospheric production (i.e. either limited by the
8 availability of nitrogen oxides NO_x or volatile organic compounds VOC).

9 In the present paper, we characterize the daily evolution of a major ozone outbreak across
10 East Asia in early May 2009, using the new multispectral satellite approach IASI+GOME2 in
11 synergism with chemistry-transport and meteorological models as well as other observations
12 (CO , NO_2 , CH_2O , etc.). We present the first observational description of the transport
13 pathways of ozone plumes from satellite measurements at the LMT (below 3 km of altitude)
14 over East Asia and we analyse the processes controlling the lowermost tropospheric ozone
15 burden during this event (i.e. photochemical production and downward transport from the
16 stratosphere). Our study uses the ratio between the enhancements of O_3 and CO to
17 characterize the Lagrangian production of ozone along transport across East Asia, derived for
18 the first time from ozone satellite data sensitive at the LMT. First, the paper presents the
19 datasets used in the study and a quality assessment of the IASI+GOME2 ozone observations
20 by comparing them with in situ measurements performed by ozonesondes and also by surface
21 stations (section 2). This comparison illustrates the unprecedented capacity of IASI+GOME2
22 to observe from space the variability of surface ozone concentrations. Section 3 describes the
23 regional distribution of ozone plumes and the meteorological conditions during each day of
24 the pollution outbreak. Then, we focus on the Lagrangian evolution of one of the major ozone
25 plumes, analysing the possible enhancement of ozone concentrations by photochemical
26 production during transport (section 4). A summary is provided in section 5.

27 **2 Datasets description**

28 **2.1 Satellite observations of lowermost tropospheric ozone: IASI+GOME2**

29 The multispectral satellite approach IASI+GOME2 is designed for observing lowermost
30 tropospheric ozone by synergism of thermal IR atmospheric radiances observed by IASI and

1 UV earth reflectances measured by GOME-2. Both instruments are onboard the MetOp
2 satellite series (in orbit since 2006 and expected until 2022) and they both offer global
3 coverage every day (for MetOp-A around 09:30 local time) with a relatively fine ground
4 resolution (12 km-diameter pixels spaced by 25 km for IASI at nadir and ground pixels of 80
5 km × 40 km for GOME-2). As described in detail by Cuesta et al., (2013), IASI+GOME2
6 jointly fits co-located IR and UV spectra for retrieving a single vertical profile of ozone for
7 each pixel. The horizontal resolution corresponds to that of IASI, using for each pixel the UV
8 measurements from the closest GOME-2 pixel (without averaging). Spectra and Jacobians in
9 the IR and UV are respectively simulated by the KOPRA (Karlsruhe Optimized and Precise
10 Radiative transfer Algorithm; Stiller et al., 2002) and VLIDORT (Vector Linearized Discrete
11 Ordinate Radiative Transfer; Spurr, 2006) radiative transfer codes. The effects of clouds and
12 aerosols are partially taken into account by iteratively adjusting offsets for each of the 7
13 spectral micro-windows (between 980 and 1070 cm⁻¹) used in the IR and effective surface
14 albedos and cloud fractions in the UV (2 micro-windows between 290 and 345 nm). Only
15 measurements with cloud fractions below 30% are used (as determined by the FRESKO
16 algorithm, Koelemeijer et al., 2001). Ozone profiles are retrieved by a constrained least
17 squares fitting method using a Tikhonov-Phillips-type regularisation (Tikhonov, 1963).
18 Constraint strengths vary with altitude and are optimised for enhancing sensitivity to
19 lowermost tropospheric ozone while keeping acceptable total retrieval errors (in the order of
20 20 % for the LMT).

21 Here, we use an updated version of the IASI+GOME2 product, with only minor changes with
22 respect to that of Cuesta et al. (2013). Ozone profiles are retrieved at the vertical grid between
23 the surface and 60 km of altitude asl (above sea level), with steps of 1 km, 2 km and 5 km,
24 respectively below 26 km asl, between 26 and 30 km asl and above. Three a priori ozone
25 profiles derived from the climatology of McPeters et al., (2007) are used, corresponding to the
26 average over 20-30°N, 30-60°N and 60-90°N, representative of tropical, mid-latitude and
27 polar conditions. These three a priori profiles are used for IASI pixels with tropopause heights
28 (determined by the temperature vertical profile) above 14 km, between 14 and 9 km and
29 below 9 km, respectively.

30 IASI+GOME2 products include vertical profiles of ozone, partial columns, averaging kernels
31 (representing sensitivity of the retrieval to the true atmospheric state), error estimations and
32 quality flags. [Since](#) 2017, global scale IASI+GOME2 retrievals are routinely produced by the

1 | French data centre AERIS and [they are](https://www.aeris-data.fr) publicly available (see <https://www.aeris-data.fr> and
2 | <http://cds-espri.ipsl.fr>).

3 | **2.1.1 Validation of IASI+GOME2 at the LMT against ozonesondes**

4 | An assessment of the quality of IASI+GOME2 for retrieving LMT ozone is presented in
5 | Figure 1 [and summarized in Table 1](#). It is based on a comparison of IASI+GOME2 retrievals
6 | and ozonesondes measurements, for the first time spread at the global scale and for all seasons
7 | during two years. We consider ozonesondes launched from 46 different sites (spread
8 | worldwide from 69°S to 83°N and 171°W to 152°E) along the years 2009 and 2010 (provided
9 | by the World Ozone and Ultraviolet radiation Data Centre - WOUDC,
10 | <http://www.woudc.org>). Vertical resolution of the ozonesonde profiles is about ~150 m and
11 | their errors are about $\pm 5\%$ (Deshler et al., 2008). Coincidence criteria are spatial co-
12 | localization [of one-degree latitude/longitude between the locations of the launching stations](#)
13 | [of the sondes and the centre points of satellite pixels](#) (as for Keim et al., 2009; Dufour et al.,
14 | 2012; Cuesta et al., 2013) and a time frame of 12 h from the MetOp-A morning overpass (at
15 | 09:30 [local time](#)). These differences in time and location induce part of the random
16 | differences between the satellite retrievals and the ozonesondes. The comparison is made for
17 | each ozonesonde with the average of collocated satellite retrievals (thus partly reducing
18 | random errors). To account for the retrieval sensitivity, we calculate “smoothed” ozonesonde
19 | measurements (indicated in Fig. 1 as “SONDE*AVK”) by interpolating at the satellite
20 | retrieval vertical grid (with 1 km-vertical resolution below 26 km), convoluting with each of
21 | the averaging kernels (AVKs) of the collocated satellite retrievals and then taking the average.
22 | Only quality-assured retrievals of IASI+GOME2 are used (discarding too high fitting
23 | residuals, cloud fraction above 30 %, aberrant retrievals of surface temperatures, ozone
24 | profiles or AVKs). After cloud screening and quality-checks, the number of sondes with
25 | coincident IASI+GOME2 data used for this comparison is 1035.

26 | The comparison at the worldwide scale shows a good agreement of IASI+GOME2 and
27 | ozonesondes in the lowermost troposphere, with a weak mean bias (-3 %), a good correlation
28 | (0.85), a very similar variability (a ratio of standard deviations of ~1.0) and a precision of 16
29 | % (estimated as the root-mean-squared difference between the two datasets, see Fig. 1a).
30 | These good results are very similar to those obtained in a first validation exercise over Europe
31 | during the summer of 2009, with practically the same correlation, precision, variability and
32 | weak bias (Cuesta et al., 2013). As this paper focuses on East Asia [\(particularly at 25-45°N](#)

1 | [110-150°E](#)), we also present the comparison for all sondes available over this region in 2009-
2 | 2010 (112 sondes after cloud screening and quality checks), launched from the 3 Japanese
3 | sites of Sapporo, Tateno (Tsukuba, near Tokyo) and Naha (Fig. 1b). In this case,
4 | IASI+GOME2 shows similarly good performance, with a weak bias (3 %), the same
5 | variability as that of sondes, a precision of 13 % and a good correlation (0.76) slightly lower
6 | with respect to the global comparison (probably partly linked to a lower variability in the
7 | measurements).

8 | **2.1.2 Capacity of IASI+GOME2 to observe near-surface ozone**

9 | An additional quality assessment is shown in Figures 2 and 3, which evaluates the capacity of
10 | IASI+GOME2 to observe near-surface ozone pollution over East Asia. IASI+GOME2
11 | retrievals at the LMT are compared with in situ measurements at the surface, from 9 stations
12 | of the EANET (Acid Deposition Monitoring Network in East Asia, <http://www.eanet.asia>)
13 | network over East Asia, [one station from the GAW \(Global Atmosphere Watch,](#)
14 | <http://www.wmo.int>) network and one station at Fukue Island (32.8°N, 128.7°E e.g. Kanaya
15 | et al., 2016) operated by JAMSTEC institute (see the location of all these stations in Figs. 3a-
16 | b). These stations are representative of background rural environment over several Japanese
17 | islands. We consider the 2 major ozone pollution events observed at the surface over Japan
18 | during the springtime 2009, respectively on 4-9 April and 4-9 May 2009 (as suggested by
19 | higher ozone surface concentrations measured by EANET/GAW/JAMSTEC). Co-localisation
20 | in time and space is assumed within ± 1 h and $\pm 1^\circ$ degree latitude/longitude, respectively.
21 | The comparison is made between the surface in situ hourly measurements for the satellite
22 | overpass time and the average of collocated satellite retrievals. Table 2 presents the results of
23 | the comparison of all coincident satellite retrievals (both for IASI+GOME2 and IASI only)
24 | and surface measurements (the 3 datasets are available for each coincidence). We consider 2
25 | sets of surface measurements in order to account for IASI+GOME2 LMT sensitivity (which
26 | peaks near 2 km asl over land): i) those corresponding to vertical gradients $\Delta O_3^{\text{surf.-2km}}$
27 | between the surface and 2 km of altitude lower than ± 10 ppb/km and ii) the whole dataset
28 | (respectively 44 and 52 coincidences). The gradient $\Delta O_3^{\text{surf.-2km}}$ is estimated from analyses of
29 | the tropospheric ozone distribution derived from the CHASER chemistry-transport model (see
30 | section 2.3). Figures 2 and 3 shows respectively the scatter of points for the case with limited
31 | $\Delta O_3^{\text{surf.-2km}}$ (similar to that for all measurements) and an illustration of the horizontal
32 | distribution of ozone satellite retrievals and surface observations. [Ozone concentrations at the](#)

1 LMT are provided as volume mixing ratios in ppb (parts per billion), calculated as the ratio of
2 LMT partial columns (in molecules per cm²) of ozone and air (in Figures 2-14 and also used
3 for CO and other partial columns).

4 Figures 2a and 3a show a good agreement between IASI+GOME2 and the ozone in situ
5 observations at the surface. To the authors' knowledge, this is the first time that such
6 agreement is found for a satellite retrieval of ozone and surface measurements. IASI+GOME2
7 observations show a fairly good correlation (up to 0.69), a mean bias of -5 %, a precision of
8 20 % (similar to the retrieval error of IASI+GOME2 at the LMT) and a similar standard
9 deviation with respect to the surface in situ measurements. Slightly lower correlation (0.63) is
10 remarked when comparing all observations (regardless $\Delta O_3^{\text{surf.-2km}}$) of the period (52 cases),
11 but the agreement remains fairly good. This is illustrated for one of the days in the Figure 3a,
12 where IASI+GOME2 clearly captures the high concentrations of the ozone plumes over the
13 Japan Sea, south Japan and the Pacific. The multispectral satellite approach is also capable to
14 observe some of the horizontal gradients within the plume, such as the relatively lower ozone
15 concentrations over the Japanese main island (60-70 ppb) with respect to higher ones over the
16 oceans (>80 ppb).

17 The uniqueness of the performance of IASI+GOME2 to retrieve near-surface ozone is put in
18 evidence by comparing the same in situ measurements with other satellite retrievals, such as a
19 single-band IASI retrieval (described in section 2.2). We use the LISA IASI product that
20 offers the highest sensitivity to ozone below 6 km (LT) among three French IASI products
21 (Dufour et al., 2012) and also largely higher than a GOME-2 only product (shown by Cuesta
22 et al., 2013). This IASI retrieval is often used to analyse ozone enhancements at the lower
23 troposphere over Europe (e.g. Eremenko et al., 2008) and East Asia (e.g. Dufour et al., 2015),
24 as also done with retrievals from OMI measurements also over East Asia (Hayashida et al.,
25 2015). The IASI LMT retrieval sensitivity peaks approximately around 3 km asl over land
26 and 4-5 km over ocean, thus 1 km higher than that for IASI+GOME2 both over land and
27 ocean (see Figures 3c-d). Figure 2b shows that the IASI only retrieval is unable to clearly
28 capture the high ozone concentrations observed at the surface, particularly those above 60
29 ppb. The scatter of points for IASI retrievals is rather flat, putting in evidence a lack of
30 sensitivity to LMT ozone also shown in the horizontal map of Figure 3b. The single-band
31 retrieval variability is much lower than that measured at the surface (the ratio of standard
32 deviations is 0.65 at most). Mean RMS differences are above 30 ppb and the correlation

1 coefficient below 0.5. On the contrary, only IASI+GOME2 does capture surface ozone
2 variations over whole range from 40 to 90 ppb (Fig. 2a) and shows a unique performance to
3 capture surface ozone variability.

4 **2.2 Other satellite observations**

5 In order to analyse the origin and evolution of ozone pollution plumes, the following
6 correlative datasets are used: CO and O₃ retrievals from IASI and NO₂ and CH₂O
7 observations derived from GOME-2 and OMI. Morning time (around 9h30 local time)
8 datasets from IASI and GOME-2 are derived from the same spectra as those used in
9 synergism by IASI+GOME2. OMI overpass occurs in the early afternoon (near 13h30 local
10 time).

11 The CO retrievals used in the present paper are derived from IASI radiances using the FORLI
12 algorithm (Hurtmans et al., 2012), from the Université Libre de Bruxelles (ULB) and the
13 Laboratoire Atmosphères, Millieux, Observations Spatiales (LATMOS). This approach uses
14 pre-calculated lookup tables of absorbance cross-sections at various pressures and
15 temperatures, and the optimal estimation for the inverse scheme. The algorithm derives
16 vertical profiles of CO, on a grid of 18 equidistant layers of 1 km of depth from the surface up
17 to 18 km, and a unique layer from 18 to 60 km. Radiative transfer calculations use operational
18 MetOp-A L2 temperature and humidity profiles, surface emissivity climatologies (Zhou et al.,
19 2011). A priori CO profiles are taken from MOZAIC, ACE-FTS for higher altitudes
20 (Clerbaux et al., 2005) and the LMDz-INCA global chemistry-transport model (Haugustaine
21 et al., 2004). FORLI provides vertical profiles, total and partial columns of CO derived by
22 profile integrations, averaging kernels, error estimations and quality flags (supplied by AERIS
23 and LATMOS). Comparisons of CO total columns derived from FORLI-IASI showed an
24 agreement better than 7 % and no significant bias with respect to other satellite products (for
25 the northern hemisphere, George et al. 2009) and ground-based retrievals from 6 NDACC
26 stations (Kerzenmacher et al., 2012). A validation of lower (surface-480 hPa) and upper (480-
27 225 hPa) tropospheric columns with respect to MOZAIC measurements found an agreement
28 of respectively 21% and 10 %, and correlations of respectively ~0.8 and ~0.7 (De Wachter et
29 al., 2012).

30 In the present study, we use CO retrievals at the LT integrated from the surface up to 6 km asl
31 (equivalent to surface-480 hPa), validated by De Wachter et al., (2012) and presenting heights

1 of maximum of sensitivity located at 3-5 km of altitude (i.e. thus at the middle of this partial
2 column, see section 4). LT partial columns are retrieved with 0.83 degrees of freedom (DOF,
3 i.e. number of independent pieces of information in the retrieved profile) in average over the
4 region and period studied in the paper. This product provides significant information on CO
5 variability below 3 km asl, as DOF at the LMT are 0.51 on average. For estimating the ratio
6 of enhancements of O₃ and CO at the LMT (hereafter referred to $\Delta O_3/\Delta CO$) during individual
7 long-range transport events, we use O₃ and CO satellite observations (mixing ratios in ppb)
8 after subtracting background levels, as done for analysing airborne in situ data during the
9 PHOBEA I and II experiments over northeast Pacific (Price et al., 2004). We empirically
10 estimate these background concentrations as the daily average concentration minus the
11 standard deviation over the region of analysis (20-48°N 110-150°E). For the event in early
12 May 2009, we derive background levels around ~46 ppb and ~126 ppb for the observations of
13 respectively LMT O₃ and LT CO. We use the same criteria for deriving $\Delta O_3/\Delta CO$ at the LMT
14 from models (WRF-Chem and CHASER, see sections 2.3 and 4).

15 We also use single-band IASI only retrievals of ozone (Eremenko et al., 2008; LISA product
16 in Dufour et al., 2012) in order to analyse the distribution of ozone between 3 to 6 km asl.
17 This single-band IASI product is similar to IASI+GOME2, but only using infrared
18 measurements (both approaches using a Tikhonov-Philips regularization and the KOPRA
19 radiative transfer code in similar configuration). By comparing IASI only and IASI+GOME2
20 ozone retrievals, one may identify ozone plumes located below 3 km asl and those located
21 between 3 and 6 km asl (as remarked by Cuesta et al., 2013). Indeed, we expect that high
22 ozone concentrations clearly depicted by IASI+GOME2 and not by IASI are located at the
23 LMT below 3 km asl. Ozone plumes located at 3-6 km asl are shown by both IASI+GOME2
24 and IASI. Note that since DOF for IASI+GOME2 is lower than 1 at the LMT, multispectral
25 outputs depend as well on ozone concentrations up to 5 or 6 km asl and they alone cannot tell
26 whether the ozone plumes are located in the LMT or at 3-6 km asl.

27 Satellite retrievals of NO₂ and CH₂O from GOME-2 (overpass around 9h30 local time) and
28 OMI (13h30 local time) are used to indicate the availability of ozone precursors. These
29 datasets are provided by TEMIS (<http://www.temis.nl>) and BIRA-IASB for CH₂O
30 (<http://h2co.aeronomie.be>). Retrievals of NO₂ from GOME-2 and OMI are derived
31 respectively by the algorithms TM4NO2A version 2.3 (Boersma et al., 2004) and DOMINO
32 version 2.0 (Boersma et al., 2011). These approaches follow 3 steps: using Differential

1 Optical Absorption Spectroscopy (DOAS) to obtain NO₂ slant columns from reflectance
2 spectra, separating the stratospheric and tropospheric contribution to the slant column and
3 converting the tropospheric slant column to a vertical column with the tropospheric air mass
4 factor. Uncertainties for tropospheric NO₂ retrievals are estimated as 35-60 % for GOME-2
5 and 25 % for OMI. Total columns of CH₂O are retrieved with the BIRA-IASB algorithm
6 version 14 (De Smedt et al., 2008), also based on DOAS technique and air mass factor
7 estimations. Uncertainties of single CH₂O slant column observations typically range from
8 about 10–200 %, when exceeding the global background ($\sim 4 \times 10^{15}$ molecules cm⁻²). For
9 reducing noise, datasets are averaged in regular grids of 1 x 1° and 2 x 2° respectively for
10 NO₂ and CH₂O.

11 **2.3 Chemistry-transport models: WRF-Chem & CHASER**

12 In the present study, we use WRF-Chem and CHASER chemistry-transport models for
13 completing the description of the ozone pollution outbreak across East Asia in early May
14 2009 and for verifying consistency with satellite observations. While WRF-Chem is a
15 regional model (Grell et al. 2005) operating in forecast mode over Asia (Takigawa et al.,
16 2007), CHASER is a global model (Sudo et al., 2002; [Sudo and Akimoto, 2007](#); [Sekiya and](#)
17 [Sudo, 2014](#)) with coarser spatial resolution but with enhanced accuracy through assimilation
18 of several satellite retrievals of atmospheric pollutants (Miyazaki et al., 2012, 2015) and with
19 higher model top height. These models provide useful insights on the detailed vertical
20 distribution (e.g. surface concentrations) and diurnal evolution of tropospheric O₃ and CO
21 over East Asia, in complement to daily satellite observations. The daily evolution of ozone
22 enhancement in the LMT is compared between the models and the satellite data. For this, we
23 estimate background levels of O₃ and CO for each dataset using the same criteria (i.e. daily
24 average minus standard deviation over the domain, see section 2.2). We derive concentrations
25 of O₃ and CO at the LMT by vertical integration from the surface up to 3 km asl (without any
26 smoothing by the satellite AVKs). Moreover, we perform sensitivity studies with CHASER
27 (in forecast mode), accounting or not for the stratospheric contribution of ozone in order to
28 identify the tropospheric or stratospheric origin of this pollutant. [In the figures of the paper,](#)
29 [we show one of the models or both of them according to the following criteria: i\) WRF-Chem](#)
30 [describes the structure of plumes of LMT O₃ and CO with finer spatial resolution \(section 3\),](#)
31 [ii\) CHASER forecasts with and without stratospheric ozone distinguish tropospheric ozone](#)
32 [formed at the troposphere from that originating from the stratosphere \(sections 3 and 4\), iii\)](#)

1 both WRF-Chem and CHASER are used for showing the temporal Lagrangian evolution of
2 O₃, CO and NO₂ for polluted air masses (section 4) and iv) CHASER analyses indicate the
3 vertical gradients of ozone between the surface and 2 km of altitude with presumably good
4 absolute accuracy provided by assimilation of various observations (section 2).

5 For the current application, WRF-Chem resolution is set to 37 vertical layers from the surface
6 up to 100 hPa (~15 km of altitude) and a Lambert conformal conic projection of horizontal
7 pixels of about ~0.9°x0.9°. Emissions from automobiles and other anthropogenic sources are
8 taken from EAgrid 2000 (East Asian Air Pollutant Emissions Grid Inventory; K. Murano,
9 personal communication) and the JCAP (Japan Clean Air Program; Kannari et al. 2007) over
10 Japan for automobile. Surface emissions over China and North and South Korea are taken
11 from REAS (Regional Emission Inventory in Asia; Ohara et al. 2007) version 1.11, and over
12 Russia from EDGAR (Emission Database for Global Atmospheric Research; Olivier et al.
13 1996) version 3.2. Biogenic emissions are based on Guenther et al. (1993). The lateral
14 boundaries of chemical species are taken from the global CHASER model every 3 hours. The
15 system is driven by meteorological data from the mesoscale model (MSM) of the Japan
16 Meteorological Agency (JMA).

17 CHASER analyses are obtained from an advanced chemical data assimilation system
18 combining satellite observations of several chemical compounds: NO₂, O₃, CO, and HNO₃
19 measurements from OMI, Tropospheric Emission Spectrometer (TES), Measurement of
20 Pollution in the Troposphere (MOPITT), and Microwave Limb Sounder (MLS). Assimilation
21 is performed according to the local ensemble transform Kalman filter technique (Hunt et al.,
22 2007), which simultaneously optimizes targeted chemical species, as well as the emissions of
23 O₃ precursors (i.e., NO_x and CO), while taking their chemical feedbacks into account.
24 Comparisons against independent data show that data assimilation results in substantial
25 improvements. It reduces biases for tropospheric NO₂ columns by 40–85 %, for lower
26 tropospheric CO concentrations in the Northern Hemisphere by 40–90 % and for O₃ in the
27 middle and upper troposphere by 30–40%. Data assimilation also mostly removed the
28 model's negative bias in surface CO concentrations in the northern hemisphere. The error
29 reduction for O₃ was generally smaller in the lower troposphere than in the middle and upper
30 troposphere because of the reduced sensitivity of the assimilated TES retrievals to lower-
31 tropospheric ozone. The CHASER forecast model includes detailed chemical and transport
32 processes in the troposphere, including 88 chemical and 25 photolytic reactions with 47

1 chemical species, and has a horizontal resolution of T42 (2.8°) and 32 vertical levels from the
2 surface to 4 hPa for the present study. Sekiya et al. (2018) have recently developed a high-
3 resolution version of CHASER with 0.56° horizontal resolution and demonstrated improved
4 performances over areas with strong local sources with respect to the 2.8° resolution version.
5 Nevertheless, the CHASER model with 2.8° resolution is capable of properly simulating
6 synoptic ozone patterns.

7 CHASER is coupled to the Center for Climate System Research/National Institute for
8 Environmental Studies (CCSR/NIES) atmospheric general circulation model (AGCM)
9 version 5.7b. The AGCM fields are nudged toward the National Centers for Environmental
10 Prediction/Department of Energy Atmospheric Model Intercomparison Project II (NCEP-
11 DOE/AMIP-II) reanalysis (Kanamitsu et al., 2002) at every time step of the AGCM to
12 reproduce past meteorological fields. Anthropogenic, biomass burning and biogenic/soil
13 emissions are respectively based EDGAR version 3.2 (Olivier et al., 2005), the Global Fire
14 Emissions Data base (GFED) version 2.1 (Randerson et al., 2007) and the Global Emissions
15 Inventory Activity (GEIA) inventory (Guenther et al., 1995). Surface emissions over Asia
16 were obtained from REAS.

17 For the sensitivity analysis, CHASER simulations of ozone “from the troposphere” (not
18 accounting for stratospheric ozone) are obtained by setting to zero ozone concentrations
19 above 100 hPa, starting from 1 January 2009. Differences between ozone concentrations from
20 full simulations and those not accounting for the stratospheric contribution provide an
21 estimation of the distribution of ozone transported “from the stratosphere”. For consistency,
22 no data assimilation is performed in either of the two simulations of this sensitivity analysis.

23 **2.4 Meteorological data: ERA-Interim reanalyses & Hysplit dispersion model**

24 Meteorological conditions leading to production of ozone pollution and transport across East
25 Asia are described in section 3 with ERA-Interim reanalyses (Dee et al., 2011) produced by
26 ECMWF (European Centre Medium Weather Forecast). We use meteorological fields
27 (downloaded from <http://climserv.ipsl.polytechnique.fr>) with global coverage, a horizontal
28 resolution of 0.75° × 0.75°, 37 pressure levels, and a time step of 6 h (interpolated for other
29 hours). Wind, geopotential height, and equivalent potential temperature fields describe
30 atmospheric circulation and the locations of synoptic high and low-pressure systems.

1 Additionally, forecasted atmospheric boundary mixing layer top heights from ERAI are used
2 in the analysis of the vertical distribution of LMT ozone derived from IASI+GOME2.

3 Trajectories of polluted air masses transported across East Asia are estimated using the
4 HYSPLIT dispersion model (Stein et al., 2015; Rolph et al., 2017, <https://ready.arl.noaa.gov>).

5 This tool can simultaneously track a total of 12500 air parcels released at a location and
6 altitude and transported during 24 hours according to meteorological fields. For the present
7 analysis, we set the starting altitudes from the surface up to 3 km (i.e. at the LMT which is the
8 layer observed by IASI+GOME2) and use built-in model reanalysis from NCEP/NCAR
9 (National Centers for Atmospheric Prediction / National Center for Atmospheric Research)
10 with a horizontal resolution of 2.5° x 2.5° and 18 (29) pressure (sigma) levels. We run the
11 dispersion model at 0 UTC (9h00 [Japan Local Time JLT](#), close to the MetOp satellite
12 overpass) [in forward mode](#) for each day of the ozone pollution event, at the mean arrival
13 location of the trajectories from the previous day. The starting point for the pollution event
14 was determined at the south of the North China plain, as suggested by model simulations and
15 high concentration of ozone precursors (see section 3.1). [For the first day, we determine the
16 region covered by the pollution plume using the dispersion model in backward mode, starting
17 at the mean location of the air parcels on the second day.](#)

18 **3 Ozone pollution outbreak across East Asia in early May 2009**

19 In this section, we describe the daily evolution of a major ozone pollution outbreak initiated
20 over China and transported across East Asia during the period 2-9 May 2009. First, we focus
21 on the formation of a large ozone plume over the North China Plain (NCP, section 3.1). Next,
22 we analyse the transport of these polluted air masses over the NCP in the northeastern
23 direction (section 3.2), and their subsequent advection by an anticyclonic circulation over
24 Northern China and the Korean Peninsula (section 3.3). Finally, these ozone plumes split into
25 two filaments and reach Japan and the Pacific far from the main sources of ozone precursors
26 (section 3.4). According to EANET/GAW/JAMSTEC surface observations over Japanese
27 islands, this is one of the two largest ozone pollution outbreaks reaching Japan during the
28 springtime of 2009.

3.1 Ozone plume formation over the NCP on 2 May

The NCP is a well-known hotspot of pollutant emissions of worldwide relevance (e.g. Richter et al., 2005). On 2 May 2009, large concentrations of ozone precursors, such as nitrogen dioxide (NO_2) and formaldehyde (CH_2O), are observed over the NCP. This is suggested by GOME-2 satellite retrievals at 34-37°N 113-117°E, marked as a magenta rectangle in Figure 4a-b. Whereas a dense NO_2 plume is mainly formed over the NCP, the highest concentrations of CH_2O are mainly located south of it (at 25-36°N 112-120°E) reaching the southern part of the NCP. The NO_2 and CH_2O concentrations observed on 2 May 2009 are approximately a factor ~ 2 higher than the regional monthly average (also estimated with GOME-2). According to MODIS active fire data over this region, wildfires are negligible in early May 2009 (only very few fire spots are detected, see <http://firms.modaps.eosdis.nasa.gov>). The short lifetimes of these reactive gases (up to a few hours) prevent the influence of long-range transport. The observed NO_2 and CH_2O plumes are then likely associated with local anthropogenic emissions from this densely populated and industrialized region. In the following (sections 3 and 4), we particularly focus our analysis on the daily evolution of pollutant concentrations originated from these air masses as they are transported across East Asia. In Figures 4-12, satellite pixels used to describe the evolution of these polluted air masses are depicted by magenta and red rectangles. These boxes contain valid satellite pixels co-located with at least 5 % of the polluted air parcels trajectories simulated by Hysplit.

According to model simulations, ozone concentrations over the NCP are relatively low (below 50 ppb) during the morning of 2 May, both at the surface and within the LMT (up to 3 km asl, shown in Figures 4c,e for WRF-Chem). Therefore, we do not observe any significant residual ozone plume from the previous day within the LMT. As expected for this time of the day (10h00 JLT), the mixing boundary layer over land is rather shallow (with its top below 1 km asl, see blue isolines representing the depth of the mixing boundary layer in Fig. 4e). Weather conditions over the region are characterized by very low windspeeds at the lower atmospheric levels associated with marked anticyclonic conditions (see high pressures approaching the NCP from the west at 25-35°E 110-115°E in Fig. 4c-d). Due to partial cloud cover, IASI+GOME2 retrievals are not available over this region during this day.

Following up with the typical diurnal cycle, ozone is photo-chemically produced during the afternoon, until reaching concentrations of ~ 90 ppb near the surface in the southern part of the NCP at 16h00 JLT (according to the WRF-Chem model around 34°N 115°E, see Fig. 4d). In

1 the afternoon, the mixing boundary layer over this region is deeply developed with its top
2 near ~2 km above ground level - agl (3.5 km asl, Fig. 4f), thus suggesting that the ozone
3 plume freshly formed during the afternoon of this day is well mixed within the LMT.

4 **3.2 Ozone plume transported over the NCP on 3-4 May**

5 In the morning of 3 May, anticyclonic conditions prevail over the NCP with a pressure
6 maximum over central China, at the southern outskirts of the NCP (see Fig. 5c). According to
7 the Hysplit dispersion model, the ozone plume formed the previous afternoon over southern
8 NCP is transported by weak southerly winds until 35°N 115°E (magenta rectangle in Fig. 5a-
9 b). At this location, high concentrations of both LMT ozone (~90 ppb) and LT carbon
10 monoxide (~290 ppb) are observed from space respectively by IASI+GOME2 (Fig. 5a) and
11 IASI (Fig. 5b). Co-localisation of both pollutant plumes over the NCP suggests that these high
12 ozone concentrations are associated with surface anthropogenic emissions, as CO is a tracer
13 for combustion-related emissions.

14 Meanwhile, another ozone plume is observed over northeastern China (north of 42°N and at
15 115-135°E in Fig. 5a). The origin of this plume is likely associated with a low-pressure
16 system (see “L” and concentric isobars north of 44°N and at 120-135°E in Fig. 5c). Such
17 systems may both entrain ozone from the stratosphere and the UTLS region down to the
18 lower troposphere and also mix pollution-related ozone within the low atmospheric levels, as
19 analysed for other events in the same region by Dufour et al. (2015). Both phenomena may
20 occur in this case. On one hand, downward transport of ozone from the stratosphere (likely
21 west of 120°E and north of 42°N) is suggested by enhanced potential vorticity at 300 hPa (a
22 tracer of stratospheric air masses) north of 48°N 130°E (Fig. 5d). On the other hand, an
23 anthropogenic contribution of LMT ozone is indicated by the presence of a CO plume
24 observed by IASI east of 122°E (and north of 40°N, Fig. 5b) likely originating from
25 northeastern Chinese emissions. This pollution plume is observed ahead of a cold front (violet
26 curve in Fig. 5c), associated with the low-pressure system north of 44°N. In this region, we
27 expect the formation of a warm conveyor belt. This ascending air stream typically mixes up
28 the air masses near the surface within the low atmospheric levels (e.g. Cooper et al., 2002;
29 Ding et al., 2009; Foret et al., 2014). Such vertical mixing likely contributes to the
30 observation of near-surface pollutants by satellite retrievals sensitive within the LMT. At this
31 location (40-45°N 122-128°E), models show ozone concentrations only enhanced up to ~50

1 ppb (Fig. 6b for WRF-Chem), near background levels. CHASER clearly simulates the
2 contribution of stratospheric ozone down to the LMT (north of 42°N), but shifted to the west
3 (95-105°E, not shown). As the paper does not focus on these air masses, a detailed analysis of
4 these differences is beyond the scope of the current paper.

5 Over the NCP (and also south of it), models simulate relatively high ozone concentrations
6 (~70 ppb) within the LMT at 10h00 JLT (see WRF-Chem in Fig. 6b). As previously
7 mentioned, IASI+GOME2 also retrieves high LMT ozone concentrations over the NCP
8 (around 35°N 115°E, Fig. 5a), but also north of it (differing from simulations at the LMT). At
9 the surface, ozone concentrations simulated by WRF-Chem remain rather low (near 40 ppb)
10 at this time of the day (Fig. 6a), probably due to titration during the previous night. This
11 suggests that the high ozone concentrations observed by IASI+GOME2 at the LMT (over the
12 NCP) likely correspond to a plume located within the residual boundary layer and formed
13 during the previous day. Such an ozone plume is simulated by WRF-Chem between 1 and 3
14 km of altitude at 35-38°N, while reaching the surface south of 35°N (Fig. 6c for WRF-Chem).
15 Both WRF-Chem and co-localisation with a plume of CO confirm the anthropogenic origin of
16 the ozone plume at the LMT observed by IASI+GOME2 over the NCP. At this continental
17 location, high NO₂ concentrations are also both observed by GOME-2 (Figs. 6d) and
18 simulated by the models (not shown).

19 The origin of this ozone plume may also be estimated from a comparison between CHASER
20 simulations in two configurations: accounting or not for the contribution of ozone from the
21 stratosphere (see more details in section 2.3). This analysis suggests that the stratospheric
22 contribution over the NCP is practically negligible (~5 ppb) at the LMT (Fig. 6e). Only above
23 the LMT (between 3 to 6 km asl), a higher contribution of ozone (~30 ppb) from a
24 stratospheric filament is depicted by CHASER along a front extending from 32°N 110°E to
25 48°N 142°E (Fig. 6f). This is also suggested by co-located high values of PV at 300 hPa from
26 ERAI (Fig. 5d). IASI-only retrievals also depict this ozone filament above 3 km asl (although
27 most pixels are cloudy, not shown).

28 During the following day (4 May), the anticyclone slowly moves northeastwards and
29 approaches the Yellow Sea, with its pressure maximum at sea level near the coast (36°N
30 122°E, Fig. 7c). According to Hysplit, the ozone plume located in the residual boundary layer
31 the previous day is advected northwards by the anticyclonic circulation up to 39°N 117°E (see
32 magenta dots in Fig. 7c). At this location over the northern part of the NCP, co-located

1 plumes of LMT ozone and CO are consistently observed respectively by IASI+GOME2 (Fig.
2 7a) and IASI (Fig. 7b). With respect to the previous day, the observed ozone concentrations
3 remain rather high (~90 ppb) while CO concentrations start dropping (~270 ppb). Over
4 northern China (north of 41°N), LMT ozone is transported eastwards following a low-
5 pressure system centred at 50°N 140°E (not shown). The stratospheric filament depicted by
6 potential vorticity at 300 hPa east of 119°E (Fig. 7d) is transported southeastwards, far from
7 the location of the ozone plumes observed at the LMT.

8 **3.3 O₃ plumes crossing Northern China and the Korean Peninsula on 5-7 May**

9 The anticyclone reaches the centre of the Yellow Sea (36°N 122°E) on the next day (5 May,
10 Fig. 8c) where it remains for two more days (until 7 May). On 5 May, the CO plume observed
11 by IASI shows an almost identical horizontal structure as the O₃ plumes seen by
12 IASI+GOME2, extending across the Yellow Sea coast from the northern part of the NCP until
13 the northern frontier of Korea. The ozone plume originating from the NCP clearly follows the
14 anticyclonic circulation surrounding the Yellow Sea, reaching the northern coast of the
15 Yellow Sea on 5 May (at 41°N 124°E, magenta rectangle in Fig. 8a).

16 As suggested by Hysplit and IASI+GOME2, the pollution plume splits into two filaments on
17 6 May (i.e. two clear pathways are depicted, Fig. 9a,c), one heading south towards the Korean
18 Peninsula as entrained by the anticyclonic circulation around the Yellow Sea and the other
19 one north of it is transported by eastwards winds to Northeast China (mentioned hereafter as
20 respectively “southern” and “northern” filaments in magenta and dotted red rectangles
21 respectively at 39°N 127°E and 43°N 130°E, Fig. 9a). The trajectories of the southern and
22 northern pollution plumes (respectively magenta and red in Fig. 9) are obtained by initializing
23 Hysplit respectively at the mean arrival location of the trajectories from the previous day and
24 2 degrees northeast from that position. The common geographical origin of the two plumes
25 (before 6 May) is confirmed by Hysplit trajectories in backward mode initiated at the location
26 of the two major ozone plumes clearly observed by IASI+GOME2 (e.g. south and north of
27 Japan two days after in Fig. 12a).

28 Pollutants over the Korean Peninsula are carried southwards by relatively strong winds
29 associated with a high and a low-pressure system respectively to the west and to the east from
30 these plumes (respectively centred at 35°N 120°E and 30°N 140°E in Fig. 9c). These co-
31 located O₃ and CO plumes show a progressive decrease in concentration with respect to the

1 previous days (down to ~220 ppb and ~70 ppb respectively for CO and O₃, Fig. 9). Such (at
2 least partly) reduction in pollutant concentrations may be induced by horizontal dilution along
3 transport away from their sources.

4 On 7 May, the southern pollution plume over Korea elongates southwards (magenta rectangle
5 at 37°N 128°E in Fig. 10a-b), with an apparent decrease in CO concentrations (~200 ppb). We
6 remark that the O₃ plumes over Korea (magenta rectangle) and the northeastern Chinese coast
7 (dotted red rectangle) are probably located below 3 km asl as they are only clearly shown by
8 IASI+GOME2 (Fig. 10a) and not by the IASI only retrieval (Fig. 10e).

9 Over the area of the northern pollution filament (over the northeastern Chinese coast),
10 enhancements of O₃ and CO concentrations are shown respectively by IASI+GOME2 and
11 IASI near 42°N 125-132°E (dotted red rectangles and dots in Figs. 10a-b). This pollutant
12 plume is observed ahead of the strong southward winds of a cold front (seen north of 42°N
13 between 115-125°E, violet curve in Figs. 10b), probably transporting freshly emitted pollution
14 (as suggested by moderately enhanced NO₂ concentrations at 43-45°N 126-132°E up to 6 10¹⁵
15 mol/cm² in Fig. 10c). These pollutants may originate from the densely populated
16 agglomeration around the Harbin megacity (43-46°N 125-127°E). This enhancement of
17 pollutant concentrations along the cold front is also clearly simulated by WRF-Chem (Fig.
18 10d for CO), although located slightly west of the plumes depicted by the satellite retrievals
19 (Fig. 10b). To account for this difference in the location of the plumes, the dotted red square
20 is shifted by 4° to the west in Fig. 10d with respect to the other panels. This freshly emitted or
21 produced pollution likely mixes with the aged pollution air masses originating from the NCP.
22 Moreover, we expect the formation of a warm conveyor belt ahead of that cold front (in violet
23 in Fig. 10b), which typically mixes up air masses in the low atmospheric levels (as also
24 remarked over this region on 3 May 2009, Fig. 5). Vertical mixing of freshly emitted
25 pollutants within the LMT may also explain the enhancements of O₃ and CO observed by the
26 satellite approaches.

27 Another ozone plume is observed south of the Yellow sea on 6-7 May by IASI+GOME2 and
28 IASI retrievals respectively at the LMT and 3-6 km asl (Figs. 9a, 10a,e). The origin of this
29 plume is probably related to downward transport from the stratosphere, as suggested by a co-
30 located PV filament at 300 hPa (green contours in Fig. 10e). CHASER simulations suggest
31 that this stratospheric ozone filament does not reach the LMT (Fig. 10f) and does not affect

1 ozone concentrations of the tracked pollution plumes over Korea and the northeastern Chinese
2 coast (rectangles).

3 **3.4 O₃ plumes transported over Japan and the Pacific on 8-9 May**

4 The high-pressure system moves southwards to the East China Sea on 8 May and then
5 eastwards until reaching the Pacific Ocean on 9 May (centred respectively at 28°N 122°E in
6 Fig. 11b and 27°N 130°E in Fig. 12b). The southern polluted air masses coming from Korea
7 (magenta rectangle) are entrained by the southwards circulation on an axis around 130°E in-
8 between the high and low-pressure systems (Fig. 11a-b). They reach Southern Japan (the
9 Kyushu island) on 8 May (31°N 130°E in Fig. 11a) and the Pacific on 9 May (27°N 130°E in
10 Fig. 12a). As only captured IASI+GOME2 (Fig. 11a) and not by IASI (Fig. 11c), the
11 moderately high O₃ concentrations over the Kyushu Island are probably located below 3 km
12 asl. Ozone and carbon monoxide concentrations at the location depicted by Hysplit (magenta
13 rectangles) are rather close to the background levels (particularly for CO).

14 The northern O₃ plume coming from the northeastern Chinese coast is transported over the
15 Japan Sea by strong counter-clockwise winds around the low-pressure system east of Japan
16 on 8 May (Fig. 11a) until reaching the Pacific southeast of Japan on 9 May (Fig. 12a). This
17 ozone plume is likely located at the LMT, as clearly depicted by IASI+GOME2 and not by
18 IASI (red rectangles respectively in Figs. 11a and 11c). According to CHASER and low PV at
19 300 hPa, we do not expect a significant contribution of stratospheric ozone reaching the LMT
20 over the Japan Sea on 8 May (Fig. 11e-f) and over central Japan on 9 May (Fig. 12e-f).
21 Therefore, these ozone plumes at the LMT are likely associated with photochemical
22 production along transport from precursors emitted over land (from northeastern China on 7
23 May and Japan on 8 May). Simulations from WRF-Chem also suggest significant
24 photochemical production of LMT ozone along transport from northeastern China to central
25 Japan and the Pacific (see section 4).

26 Downward transport of stratospheric ozone occurs southeast of Japan on 8-9 May, as
27 suggested by the location of the PV filament at 300 hPa (Figs. 11e, 12e), which travels
28 eastwards (located south of the Yellow Sea on 7 May). This ozone plume originating from the
29 stratosphere reaches the lower troposphere above 3 km asl (as observed by both
30 IASI+GOME2 and IASI in Figs. 11a,c, 12a,c) but not the LMT (indicated by CHASER in
31 Figs. 11f and 12f). This suggests distinct origins and vertical locations for the two elongated

1 ozone plumes southeast of Japan observed by IASI+GOME2 on 9 May (27-40°N 132-141°E
2 in Fig. 12a). The one closer to Japan is associated with photochemical production and other
3 one with stratospheric transport (indicated by green contours of PV in Fig. 12e), respectively
4 located at the LMT and 3-6 km asl.

5 **4 Photochemical production of lowermost tropospheric ozone during** 6 **transport**

7 According to the previous section, the major ozone outbreak initiated over the NCP in the
8 afternoon of 2 May 2009 is transported northeastwards over Northern China; it splits into two
9 pollution filaments and then heads southwards until reaching southern Japan on 9 May 2009.
10 Detailed analyses of the transport pathways of this large pollution plume (highlighted in
11 magenta/dotted red rectangles in Figs. 4-12) suggest two significant contributions of ozone
12 precursors from the NCP (2 May) and Northeastern China (northern pollution filament on 7
13 May). We do not observe any significant contribution of ozone from the stratosphere co-
14 located with these ozone plumes nor mixing with large ozone plumes formed in other regions.
15 In absence of local production (which occurs on 2 May and for the northern plume on 7 May),
16 we expect the evolution of LMT ozone to be mainly driven by either along-transport
17 production (linked with the availability of ozone precursors and solar insolation) or dilution of
18 the air masses (due to horizontal wind divergence and/or vertical mixing).

19 Figures 13 and 14 present a quantitative analysis of the Lagrangian evolution of the air
20 masses travelling on 3-9 May 2009 across East Asia from the NCP to southern Japan. These
21 time series show the daily evolution of a given variable averaged at the location of the
22 highlighted major pollutant plume, as depicted by the Hysplit dispersion model
23 (magenta/dotted red rectangles in Figs. 4-12), each day during the morning (at the time of
24 | overpass of the MetOp-A satellite around 9h30 local time). The key variable to analyse is the
25 ratio $\Delta O_3/\Delta CO$ that describes the relative production or decrease of O_3 with respect to CO
26 along transport (e.g. Price et al, 2004) and also used at given fixed locations (e.g. Chin et al.,
27 1994). Figures 13a-c show two curves, one corresponding to the beginning of the event and
28 the southern filament of pollution and the other for the northern pollution filament (i.e. curves
29 respectively in magenta and dotted red).

4.1 Southern pollution plume

For the southern filament, a sustained increase of the ratio $\Delta O_3/\Delta CO$ is clearly derived during the whole ozone outbreak across East Asia from satellite observations (Fig. 13a). It evolves from ~ 0.25 over the NCP on 3 May to ~ 0.46 on 8-9 May over the Pacific and southern Japan (in magenta in Fig. 13a). As dilution of the air masses affects equally the concentrations of both pollutants, a monotonous enhancement of $\Delta O_3/\Delta CO$ clearly puts in evidence the production of O_3 along transport. The evolution of this ratio from 0.25 over the main source regions to 0.46 after long-range transport estimated here with satellite retrievals is consistent with other estimations from airborne in situ measurements ranging from 0.2 to 0.5 (from flights at 2-3 km of altitude) for other events of transpacific long-range transport of industrial/urban pollution and in absence of stratospheric intrusions (Price et al., 2004). Overall consistency is also found with model estimates of 0.3 for typical air masses downwind from Asian pollution sources in springtime (Mauzerall et al., 2000) and the same value from in situ ground-based measurements over the United States in summer (Chin et al., 1994). These IASI+GOME2- O_3 /IASI-CO values are fairly higher than those estimated in the lower troposphere with IASI- O_3 only retrievals (0.16-0.28 for the column below 6 km asl) for an Eastern Asian pollution event in May 2008 (Dufour et al., 2015) and lower than retrievals in the free troposphere (400-700 hPa) from OMI/AIRS (~ 0.6) over Tokyo (Kim et al., 2013).

Assuming that most CO is emitted at the beginning of the event (for the southern filament, magenta curve in Fig. 13a), the evolution of the $\Delta O_3/\Delta CO$ values suggests a production of O_3 along transport of $\sim 60\%$ after the first 3 days (mainly over Northern China and Korea) and of $\sim 84\%$ during the whole event (6 days) with respect to that over the NCP. In this case, the greatest growth of $\Delta O_3/\Delta CO$ occurs on 3-6 May when the air masses are transported over the most industrialized areas (the NCP and Northern China) with the greatest emissions of ozone precursors as NO_x (as shown in Fig. 10c for NO_2). In the following days, a slower growth with almost constant $\Delta O_3/\Delta CO$ occurs over the Korean Peninsula (on 7 May), southern Japan (8 May) and the Pacific (9 May), far from the main sources of O_3 precursors over China. Less ozone production over this oceanic region is well consistent with low availability of NO_2 indicated both by satellite observations and both models (Fig. 14j) and a regime of NO_x -limited photochemical production of O_3 , as observed over the Fukue Islands (Kanaya et al., 2016). This behaviour is also simulated by WRF-Chem and CHASER, showing ozone diurnal cycles with greater ozone production in the afternoon of the first 3 days of the event and

1 | significantly less after (see hourly outputs of the models in Fig. 14a,c). Simulated diurnal
2 | cycles of ozone also reveal the strong nocturnal reduction in ozone concentrations (down to
3 | 30-40 ppb for WRF-Chem and 40-60 ppb for CHASER, dotted light colour curves in Fig.
4 | 14a,c) particularly significant over China (3-6 May), probably associated to nitrogen
5 | monoxide (NO) titration over the continent, and less pronounced after, near or over the ocean.
6 | Moreover, WRF-Chem clearly suggests a reduction in CO concentrations (also observed by
7 | IASI in the lower troposphere), particularly significant after 6 May (Fig. 14b) and likely
8 | linked to atmospheric dilution (horizontal and/or vertical). Sinks of CO are not expected to be
9 | significant during a period of 3 days.

10 | The $\Delta O_3/\Delta CO$ ratios derived from the CHASER and WRF-Chem models at the LMT follow a
11 | similar relative evolution as that from satellite retrievals, with a minimum at the beginning of
12 | the event and a relative monotonous increase by the end (Fig. 13b-c). This is particularly
13 | observed for WRF-Chem and until 6 May for CHASER. In absolute values, the ratio
14 | $\Delta O_3/\Delta CO$ derived from the satellite measurements is higher than that from models. At the
15 | beginning of the event (3-5 May), satellite estimates of the ratio are 0.1 to 0.15 higher than
16 | those from models. After 6 May, satellite and WRF-Chem ratios are closer (with differences
17 | between 0.05 and 0.1). Differences between the models and with respect to satellite-derived
18 | $\Delta O_3/\Delta CO$ ratios are likely associated with photochemical schemes in the models, model
19 | resolutions, precursors availability, the location of the plumes, etc. The non-steady
20 | enhancement of $\Delta O_3/\Delta CO$ for CHASER after 6 May could be partly associated with
21 | significantly less availability of NO_2 at the LMT, as compared to WRF-Chem (a factor 3
22 | higher, Fig. 14i) and a less marked reduction of CO concentrations at the LMT (while it
23 | clearly decreases for WRF-Chem, Fig. 14b,d).

24 | Figures 14g-h show that the steady increase of $\Delta O_3/\Delta CO$ observed for the southern pollution
25 | plume does not seem to be linked to changes in sensitivities of the satellite retrievals. This is
26 | described in terms of the degrees of freedom and the altitude of maximum sensitivity of the
27 | retrieved atmospheric columns, which respectively quantify the amount of information
28 | provided by the satellite retrieval and the altitude it comes from. Neither of these two
29 | variables for either O_3 and CO reflect such a steady variation, greater during the first 3 days
30 | and nearly flat afterwards, as that observed for $\Delta O_3/\Delta CO$. The LMT O_3 retrieval sensitivity
31 | peaks between 2.5 and 3 km asl for most of the days (and near 4 km asl over oceanic cold
32 | waters on 9 May), with degrees of freedom fluctuating from 0.2 to 0.3 (for the LMT, Fig. 14g,

1 | and around 5.5 to the O₃ total column, not shown). The CO lower tropospheric column is
2 | retrieved with 0.8 to 1 degrees of freedom with a peak of sensitivity from 3.5 to 5 km of
3 | altitude (Fig. 14g-h).

4 | Figure 14 (e-f, i) show evidence of the negligible influence of stratospheric ozone on the
5 | evolution of $\Delta O_3/\Delta CO$ at the LMT for the polluted air masses tracked in 3-9 May. Ozone
6 | amounts within the LMT originating from the troposphere are a factor ~ 12 greater than the
7 | contribution from the stratosphere, according to CHASER simulations (accounting or not for
8 | stratospheric contributions, Fig. 14e). This is consistent with meteorological tracers of
9 | stratospheric air masses, as the potential vorticity at 500 hPa (Fig. 14i). At this pressure level,
10 | no particular enhancement of potential vorticity is clearly remarked in correlation with the
11 | days of high concentration of ozone at the LMT. On the other hand, the ozone contribution of
12 | stratospheric downward transport at the upper troposphere (from 6 to 12 km asl) fluctuates
13 | significantly during the whole event (Fig. 14f). Potential vorticity on 3 May 2009 is high only
14 | at 300 hPa in consistency with an ozone enhancement at the upper troposphere (Fig. 14e), but
15 | not below (see potential vorticity at 500 hPa in Fig. 14i). As quality check, we remark that
16 | similar concentrations of ozone at the LMT and the upper troposphere are retrieved by
17 | IASI+GOME2 and simulated by CHASER (adding contributions from the Troposphere and
18 | Stratosphere in) averaged over the whole event (differences of 13 ppb at most).

19 | **4.2 Northern pollution plume**

20 | For the northern pollution plume, satellite-derived $\Delta O_3/\Delta CO$ ratios show an increase on 6
21 | May (curve red in Fig. 13a) with respect to the previous days, as remarked for the southern
22 | plume. On 7 May, the eastern plume airmasses exhibit lower $\Delta O_3/\Delta CO$ ratios of ~ 0.25 ,
23 | probably due to mixing with freshly emitted pollutants from the Northern China megacities
24 | (suggested by CO observations on 7 May, Fig. 10b, and NO₂ concentrations from WRF-Chem
25 | and CHASER in Fig. 14j). This value of $\Delta O_3/\Delta CO$ is practically the same as the one
26 | observed on 3 May over large pollution sources from NCP. From 7 to 9 May, the $\Delta O_3/\Delta CO$
27 | ratio (in red) raises up monotonically from ~ 0.25 to ~ 0.4 , thus suggesting photochemical
28 | production along transport (as remarked for the days followed emission of ozone precursors
29 | over the NCP). This evolution in terms of $\Delta O_3/\Delta CO$ ratios corresponds to an ozone
30 | production of about $\sim 60\%$ with respect to that on 7 May, within 2 days. The relative evolution
31 | of satellite-derived $\Delta O_3/\Delta CO$ ratios is consistent with WRF-Chem simulations (blue curve in

1 Fig. 13b), which also shows a relative increase from 5 to 6 May and then lower values on 7
2 May (with an additional pollution plume) that rise up monotonically until 9 May. The
3 CHASER model shows an enhancement from 7 to 8 May, but it drops on 9 May (Fig. 13c).
4 The latter might be linked to low availability of NO₂ at the LMT in CHASER simulations (a
5 factor 3 lower than for WRF-Chem, Fig. 14j) and eventually a difficulty to represent such
6 small-scale pollution plumes with the coarser resolution of this global model.

7 During this period, air masses are transported from Northeastern China to the Japan Sea, then
8 over Central Japan and finally reaching the Pacific. Ozone precursors might originate from
9 Northeastern Chinese and Central Japanese megacities. Both WRF-Chem and CHASER
10 simulations suggest a relatively higher availability of NO₂ at the LMT (although 3 times
11 higher for WRF-Chem) for the northern pollution filament (dotted curves with respectively
12 blue ovals and green stars in Fig. 14j) as for the southern plume (light blue and light green in
13 Fig. 14j). This is consistent with the greater growth of $\Delta O_3/\Delta CO$ (and therefore ozone
14 production) from 7 to 9 May for the northern pollution plume with respect to that at the South,
15 as estimated with satellite retrievals (Fig. 13a). WRF-Chem and CHASER simulations also
16 suggest the occurrence of ozone production along transport after the 7 May by a succession of
17 marked diurnal cycles of ozone with greater amounts in the afternoon (Fig. 14a,c). As
18 compared to the period before 6 May, ozone diurnal cycles simulated by both models exhibit
19 smaller amplitudes, which are likely associated with less nighttime titration over non-
20 continental areas. The reduction of this ozone reservoir may also enhance the growth of
21 $\Delta O_3/\Delta CO$ along transport.

22 As for the southern plume, stratospheric contribution of ozone down to the LMT at the
23 location of the northern pollution filament seems negligible according to CHASER
24 simulations (Fig. 14e) and low values of potential vorticity (Fig. 14i). Besides, satellite-
25 derived $\Delta O_3/\Delta CO$ ratios may be affected by changes in sensitivity for the CO IASI retrievals
26 that peaks at the middle troposphere on 7-8 May, instead of the lower troposphere (Fig. 14h).
27 According to sensitivity analyses, these uncertainties induce under or overestimations for
28 $\Delta O_3/\Delta CO$ that remain below $\pm 11\%$ for changes of 1 and 3 km in the heights of maximum
29 sensitivity for respectively O₃ and CO retrievals. These estimations are obtained using typical
30 vertical profiles of O₃ and CO for a pollution plume (from WRF-Chem) smoothed with
31 averaging kernels of the satellite retrievals and taking into account the concomitant change in
32 the heights of maximum sensitivity for O₃ and CO retrievals, as in both cases they depend on

1 the difference between surface and air temperatures. These uncertainties are significantly
2 lower than changes observed for $\Delta O_3/\Delta CO$ from satellite retrievals (up to 84% during the
3 whole event). Therefore, conclusions drawn on the occurrence and quantification of
4 photochemical ozone production in this period are not significantly affected by changes in
5 satellite retrievals sensitivities.

6 **5 Summary and conclusions**

7 We have presented a detailed study of the daily evolution of lowermost tropospheric ozone
8 during a major pollution outbreak across East Asia in early May 2009, by means of
9 IASI+GOME2 multispectral satellite observations and chemistry-transport models. This new
10 multispectral satellite approach offers the currently unique capacity to observe the ozone
11 distribution at the lowermost troposphere (below 3 km asl) with a maximum of sensitivity
12 down to 2 km asl over land. Comparison with respect to ozonesonde measurements show a
13 good performance of IASI+GOME2 to retrieve ozone at the LMT, in average for 46 locations
14 in all continents around the world and for all seasons (mean bias of ~3%, correlation of 0.85
15 and mean precision of 16%) and also particularly over East Asia (where the present analysis
16 is focused). Comparisons with surface in situ measurements illustrate as well the very good
17 performance of IASI+GOME2 to observe ozone pollution from space. Contrary to IASI
18 alone, IASI+GOME2 is capable of observing the spatiotemporal variability of surface ozone
19 during the 2 main pollution events in springtime 2009 over the Japanese Islands, with
20 relatively low bias (5%) and a fair correlation (0.69).

21 Using IASI+GOME2, we describe the transport pathways and daily evolution of the ozone
22 pollution outbreak in the lowermost troposphere across East Asia in early May 2009, with
23 unprecedented observational detail. We document the transport pathways of lowermost
24 tropospheric of ozone and carbon monoxide plumes from the North China Plain to the Pacific,
25 surrounding the Yellow Sea and passing over Korea and Japan. Model simulations suggest
26 that these plumes are formed near the surface on 2 May, mixed within the mixing boundary
27 layer over the lowermost troposphere (up to 3 km asl) during the day and then transported as a
28 residual boundary layer in the following days until the Pacific on 9 May. Satellite retrievals
29 depict clearly concomitant structures of LMT O_3 and CO plumes mostly every day, thus
30 suggesting the anthropogenic origin of both pollutants. Within the pollution plumes, LMT O_3
31 mixing ratios range from ~90 ppb at the beginning of the event to ~70 ppb at the end. During

1 the event, ozone concentration is affected simultaneously by both photochemical production
2 within transported air-parcels and horizontal/vertical dilution associated with atmospheric
3 circulation. We estimate that the contribution of photochemical production is an increase of
4 up to 84 % of ozone amounts with respect to that produced on the first day of the event over
5 NCP. This estimation uses O₃ to CO enhancements ratios with respect to background, for the
6 pollution plumes transported across East Asia. The evolution of this ratio is influenced by
7 sources or sinks of pollutants and not by atmospheric dilution, as this last one affects equally
8 both pollutants. This type of results represents a strong benchmark for atmospheric pollution
9 models. It has been shown, that the two models used here (CHASER, and WRF-Chem) are
10 able to reproduce the broad features of the temporal evolution of the enhancement ratio.
11 Absence of stratospheric ozone contributions confirms the photochemical origin of O₃
12 enhancements with respect to those of CO. Moreover, detailed tracking of pollution plumes
13 suggests that it splits into two pollution filaments when crossing over Northeastern China.
14 One of them is mixed with freshly emitted pollutants, with significant photochemical
15 production of ozone, but the other one follows a rather constant evolution of the O₃ to CO
16 enhancements ratio until reaching the Pacific.

17 The present satellite based approach has shown original and air-quality relevant skills to
18 describe the evolution of transboundary pollution outbreaks across East Asia. Particularly,
19 distinguishing photochemical production along transport to that originally produced over
20 major pollution sources is a significant contribution for a better understanding of air quality
21 degradation and developing efficient pollution mitigation policies. Future studies will extend
22 the approach to longer time periods and consider multiple meteorological regimes propitious
23 for East Asian pollution.

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1 **Table 1.** Validation of IASI+GOME2 ozone retrievals at the LMT against ozonesondes
2 measurements from 46 stations distributed worldwide (and over East Asia in the region 25-
3 45°N 110-150°E, i.e. 3 Japanese stations) launched in 2009 and 2010, during all seasons. We
4 account for the satellite retrieval sensitivity by smoothing ozonesonde profiles with averaging
5 kernels of IASI+GOME2 pixels with centres co-located within +/- 1° of latitude and longitude
6 from the station launching the sonde. Ozonesonde-derived LMT ozone columns are calculated
7 by vertical integration and compared with the averaged of IASI+GOME2 co-located
8 retrievals. Biases and RMS differences are given in Dobson Units (DU) and percentage in
9 parenthesis. Scatterplots of these datasets are provided in Fig. 1.

10

| IASI+GOME2 retrievals at the LMT vs. ozonesondes | Ozonesondes distributed worldwide | Ozonesondes over East Asia |
|--|-----------------------------------|----------------------------|
| Bias | -0.31 (-3.1%) | 0.37 (3.3%) |
| Correlation | 0.85 | 0.76 |
| RMS difference | 1.62 (16%) | 1.43 (13%) |
| Standard deviation ratio | 1.01 | 1.00 |
| Number of ozonesondes | 1035 | 112 |

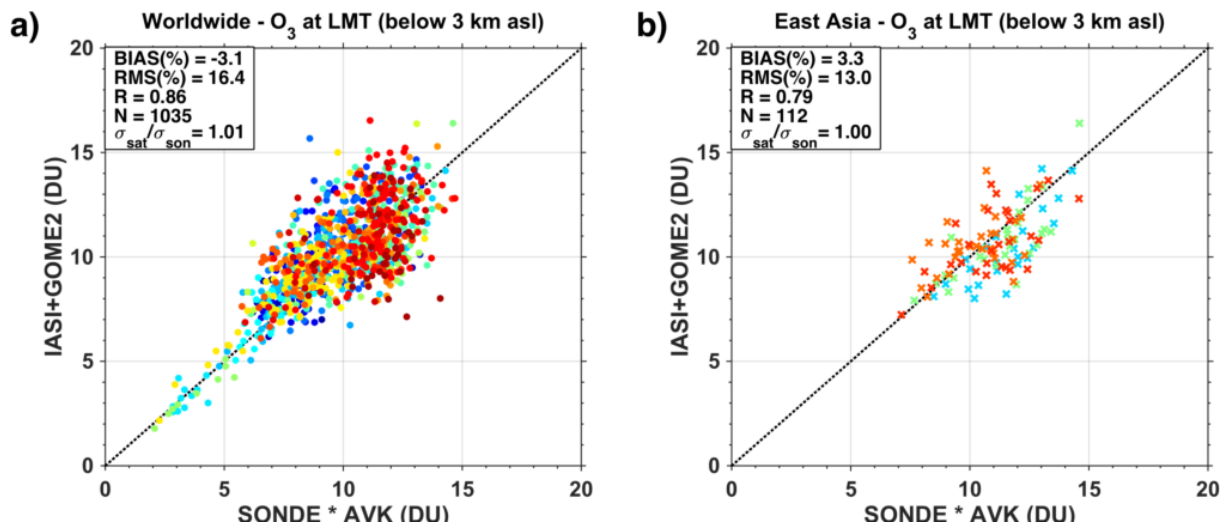
11

1 **Table 2.** Comparison of ozone in situ measurements at the surface from 11 EANET stations
2 over the Japanese islands with IASI+GOME2 and IASI only retrievals at the LMT, for two
3 major ozone outbreaks on 4-9 April and 4-9 May 2009. We consider in situ measurements at
4 10h00 [Japan Local Time \(JLT\)](#) and the average of co-located satellite retrievals +/- 1° of
5 latitude and longitude. We only account for coincidences with both IASI+GOME2 and IASI
6 retrievals. Biases and RMS differences are given in ppb mixing ratio and percentage in
7 parenthesis. Scatterplots of these datasets are provided in Fig. 2. A selection of the data with
8 limited gradient (lower than [10 ppb/km](#) in absolute value) of ozone between the surface and 2
9 km (according to CHASER [analyses](#)) is considered.

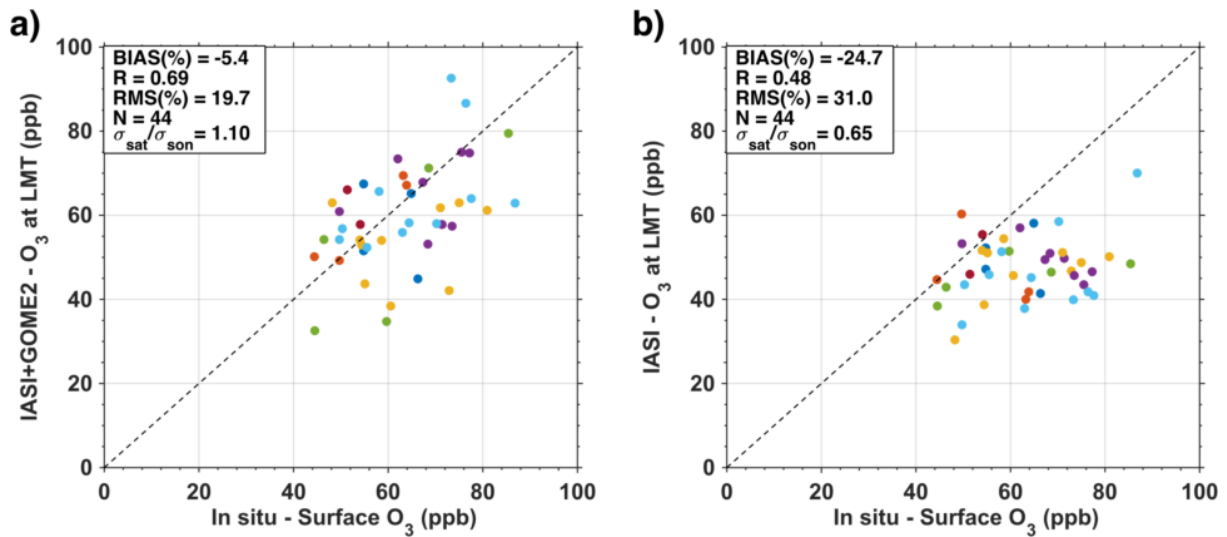
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| | IASI+GOME2 vs. Surface measurements | | IASI vs. Surface measurements | |
|-----------------------------|--|---------------|----------------------------------|-----------------|
| | Limited gradient surface-2km | All cases | Limited gradient surface-2km | All cases |
| Mean bias | -3.4 (-5.4 %) | -3.0 (-4.8 %) | -15.6 (-24.7 %) | -15.6 (-24.7 %) |
| Correlation R | 0.69 | 0.63 | 0.48 | 0.46 |
| RMS difference | 12.4 (19.7 %) | 13.5 (21.3 %) | 19.5 (31.0 %) | 20.0 (31.8 %) |
| Standard deviation ratio | 1.10 | 0.97 | 0.65 | 0.57 |
| Number of measurements | 44 | 52 | 44 | 52 |

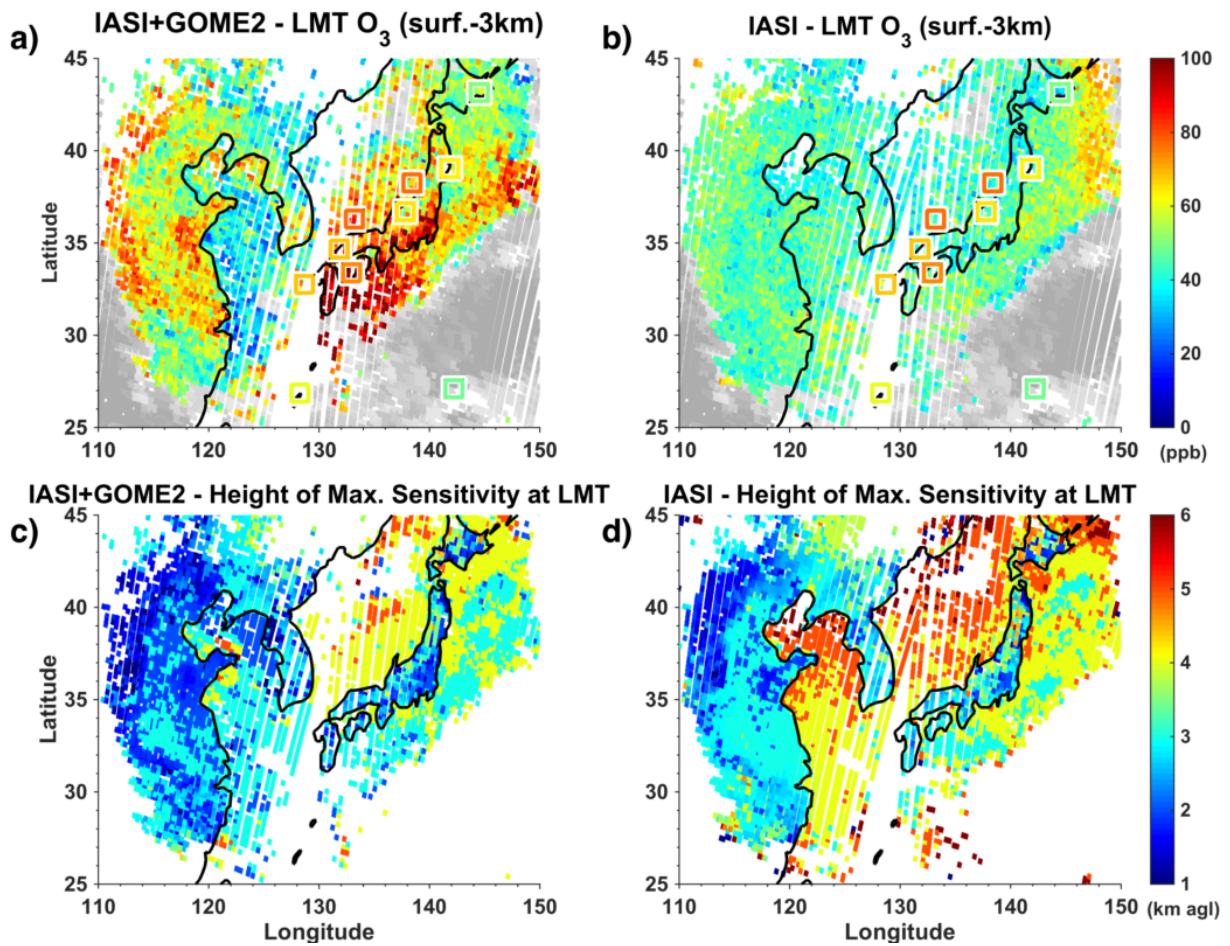
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1
 2 **Figure 1.** Validation of IASI+GOME2 retrieval of O₃ (in Dobson Units, DU) at the LMT
 3 (between the surface and 3 km asl) by comparison with ozonesondes during 2009 and 2010
 4 launched from **(a)** 46 stations spread worldwide and **(b)** 3 Japanese stations (Sapporo, Tateno
 5 and Naha) in the region 25-45°N 110-150°E. Averaging kernels of IASI+GOME2 are used
 6 for smoothing ozonesonde measurements for accounting for satellite retrievals sensitivity.
 7 The symbol $\sigma_{\text{sat}}/\sigma_{\text{sonde}}$ is the ratio between the standard deviations of the sonde data and the
 8 satellite retrievals.

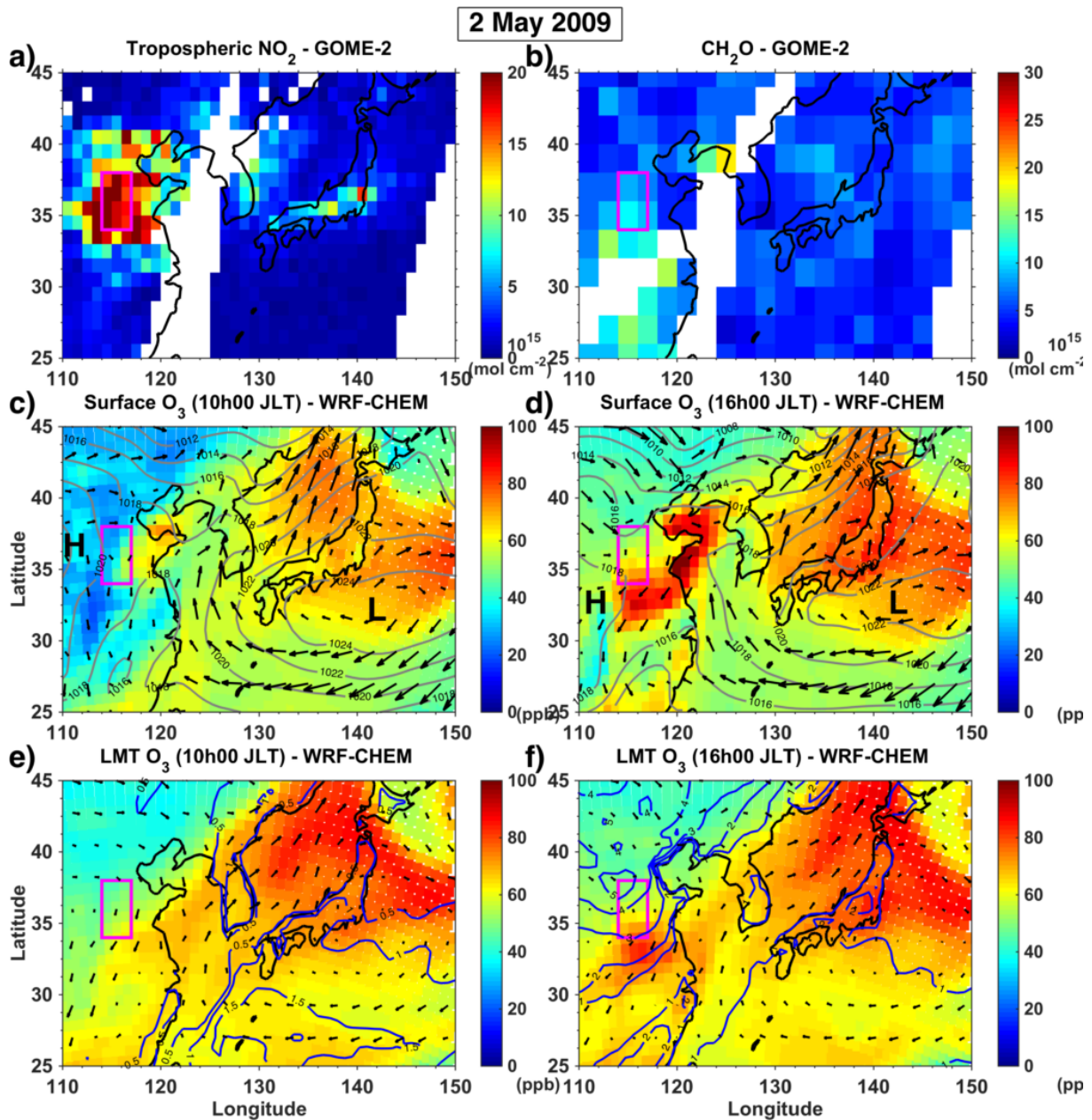


1
2 **Figure 2.** Evaluation of the capacity of IASI+GOME₂ to retrieve near-surface ozone:
3 Comparisons of **(a)** IASI+GOME₂ and **(b)** IASI only retrievals with surface ozone
4 observations from 11 EANET/GAW surface in situ stations over East Asia, during the two
5 greatest East Asian ozone pollution events in springtime 2009 (from 4 to 9 April and from 4
6 to 9 May 2009). The figures show cases with vertical gradient of ozone concentration
7 between the surface and 2 km of altitude below 10 ppb/km (according to CHASER model
8 analysis). This is a direct comparison without smoothing by averaging kernels. Colours
9 indicate different days of the comparison. The symbol $\sigma_{\text{sat}}/\sigma_{\text{sonde}}$ is the ratio between the
10 standard deviations of the sonde data and the satellite retrievals. Ozone concentrations at the
11 LMT are provided as volume mixing ratios in ppb, calculated as the ratio of LMT partial
12 columns (in mol/cm²) of ozone and air (in Figures 2-14 and also used for CO and other partial
13 columns).

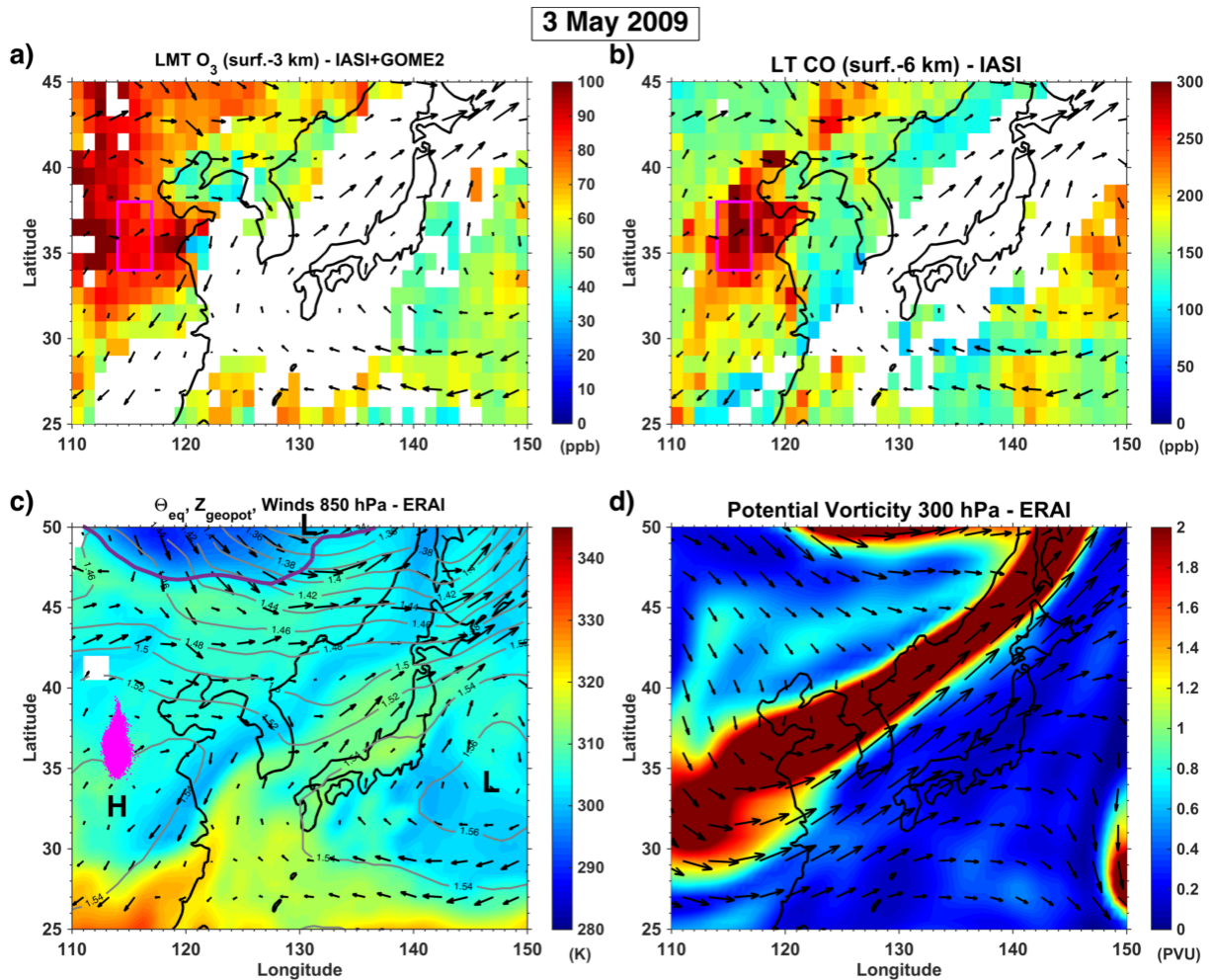


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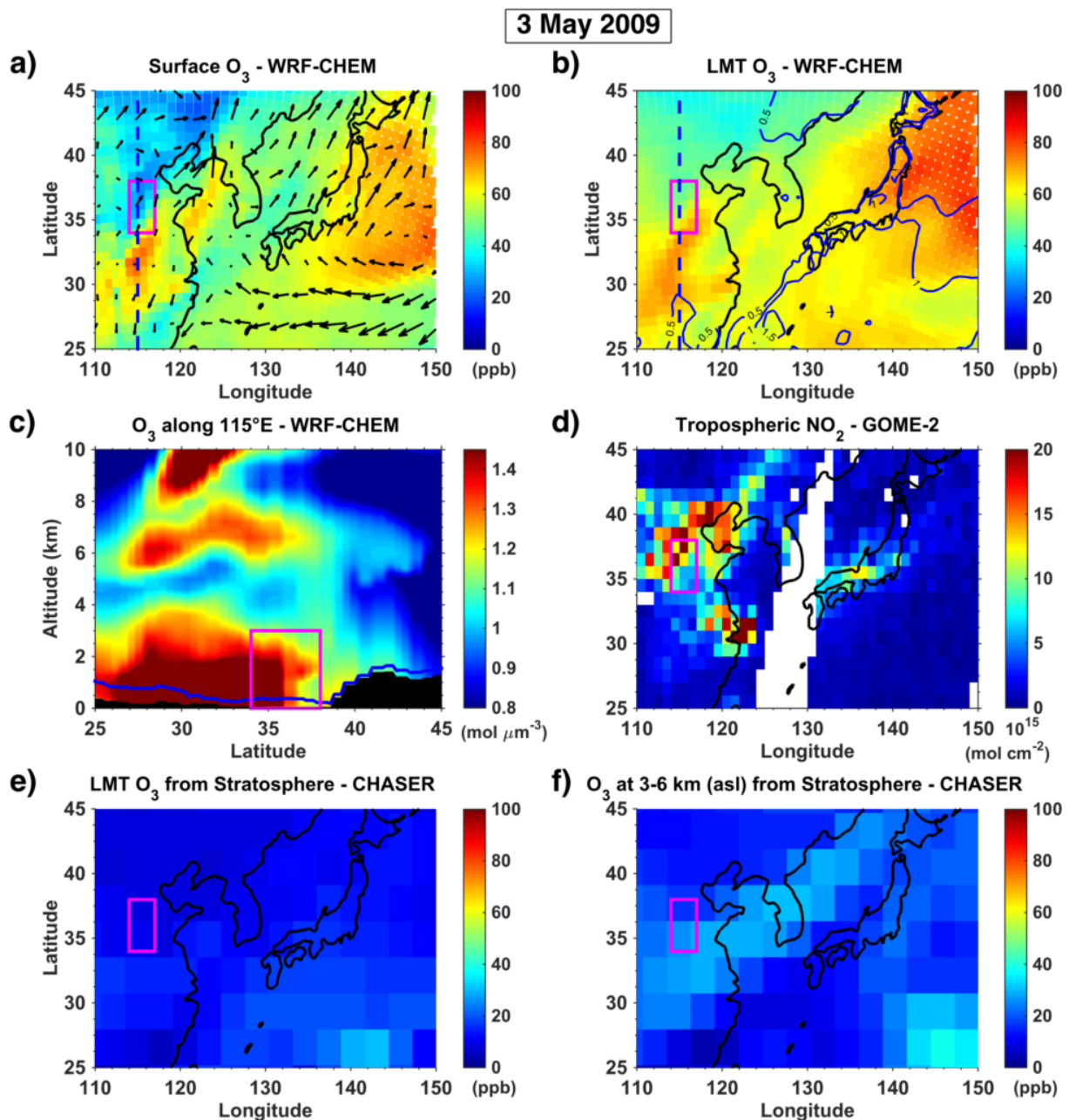
2 **Figure 3.** Example of comparison on 9 April 2009 over East Asia of (a) IASI+GOME2 and
 3 (b) IASI retrievals of LMT ozone (from the surface up to 3 km asl in both cases) with surface
 4 observations (squares in panels a and b). Grey-shaded pixels show cloud fractions above 0.3,
 5 as derived from GOME2 Fresco algorithm. Heights of maximum sensitivity of the LMT
 6 ozone partial columns are shown for (c) IASI+GOME2 and (d) IASI.



1
 2 **Figure 4.** (a) Tropospheric nitrogen dioxide NO_2 and (b) Formaldehyde CH_2O distribution
 3 over East Asia on 2 May 2009 derived from GOME-2 observations at 9h30 JLT. (c-f) Ozone
 4 distribution simulated by the WRF-Chem model at the surface (panels c and d) and averaged
 5 below 3 km of altitude agl (LMT, panels e and f), in the morning (at 10h00 JLT, panels c and
 6 e) and on the afternoon (at 16h00 JLT, panels d and f). Iso-contours in grey (panels c-d) and
 7 dark blue (e-f) are mean sea level pressure (in hPa) and mixing boundary layer height (in km
 8 asl) from ERAI reanalysis. Arrows depict winds at the surface (c-d) and 850 hPa (e-f) from
 9 ERAI. Low and high-pressure systems are indicated by respectively “L” and “H” (in panels c-
 10 d). Magenta rectangles in panels indicate the overall location on 2 May 2009 of the tracked
 11 airmasses during the ozone pollution outbreak of early May 2009.

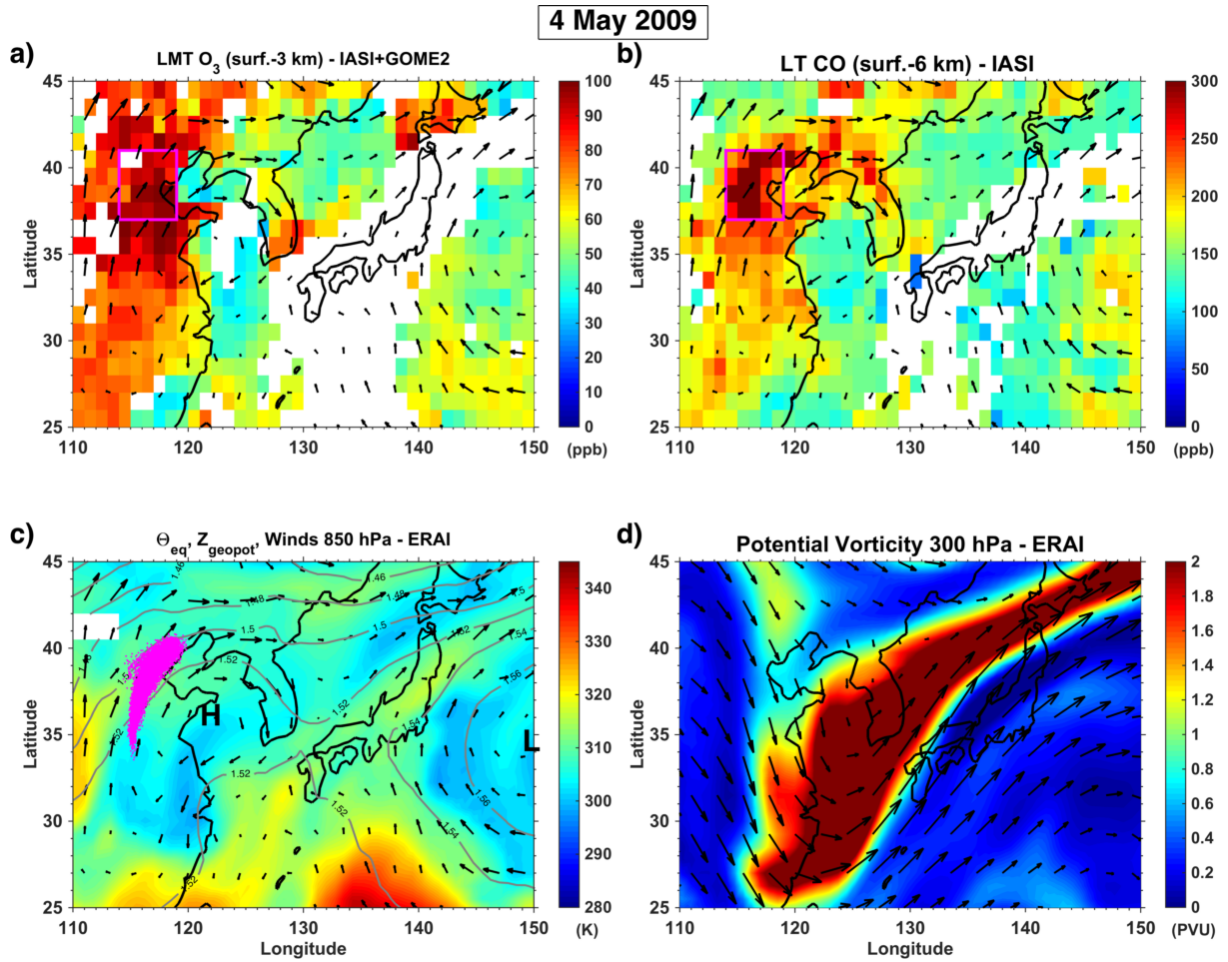


1
 2 **Figure 5.** Gaseous pollutants distributions and meteorological situation over East Asia on 3
 3 May 2009: **(a)** Lowermost tropospheric ozone derived below 3 km of altitude asl from
 4 IASI+GOME2, **(b)** Carbon monoxide at the lower troposphere (below 6 km of altitude)
 5 retrieved from IASI measurements, **(c)** Equivalent potential temperature θ_{eq} (colour shading
 6 in K), geopotential height Z_{geopot} (grey isolines every 20 m), winds at 850 hPa from ERAI
 7 reanalyses, **(d)** Potential vorticity (colour shading in PVU) and winds at 300 hPa from ERAI.
 8 The magenta rectangle in panels (a) and (b) indicate the overall location on 3 May 2009 of the
 9 tracked airmasses during the ozone pollution outbreak of early May 2009 and the magenta
 10 dots in (c) correspond to the precise air parcels locations provided by Hysplit. The location of
 11 a cold front is shown by a violet curve in panel (c).

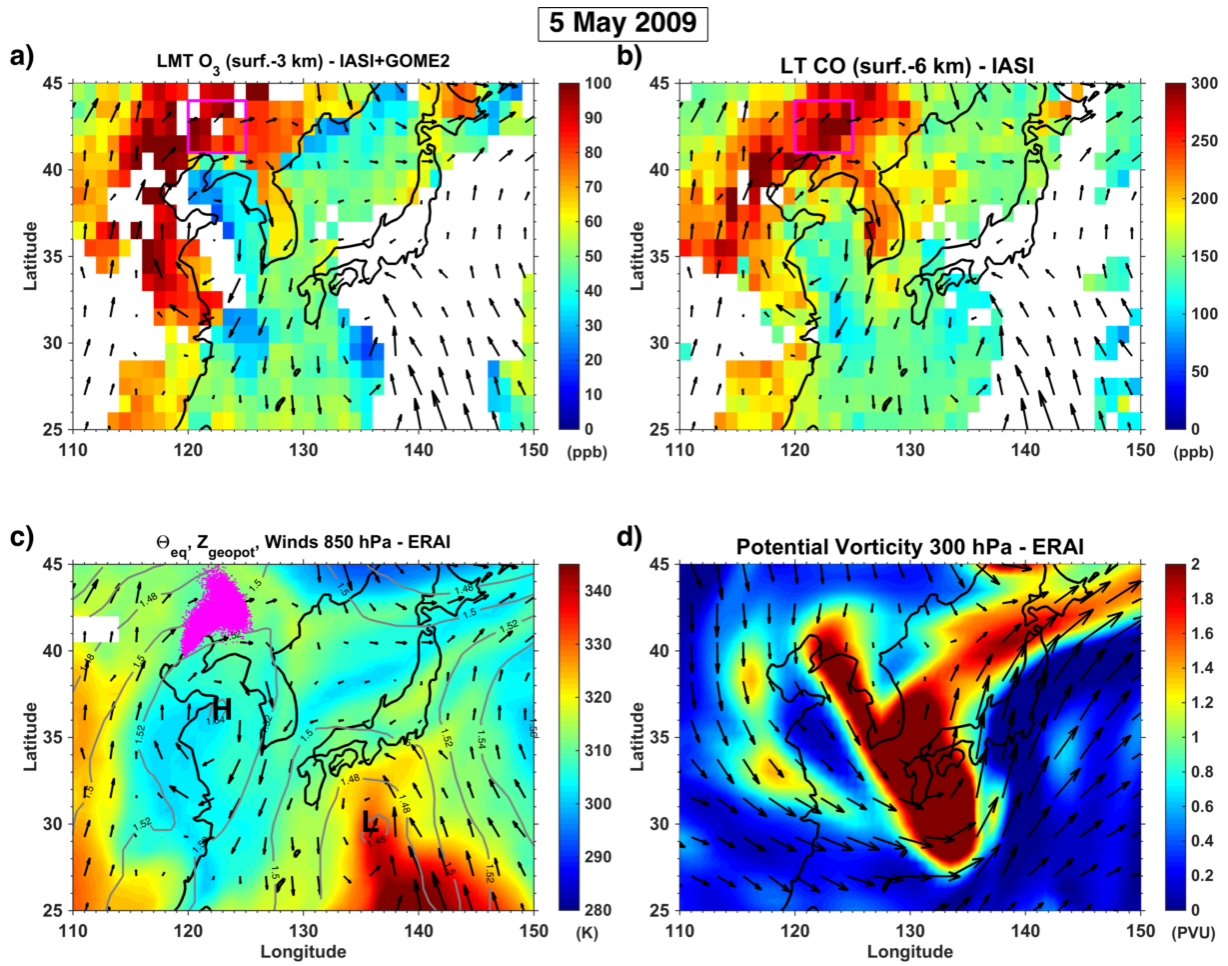


1
 2 **Figure 6.** Ozone distribution over East Asia (a) at the surface and (b) at the LMT according
 3 to the WRF-Chem model on 3 May 2009 at 10h00 JLT. Panels (a) and (b) also show surface
 4 winds (arrows) and mixing boundary layer height (blue contours), respectively. (c) Transect
 5 of vertical profiles of tropospheric ozone burden (in molecules per μm^3 of air) along the axis
 6 115°E (indicated as a dashed blue line in panel a) from derived from WRF-Chem, with the
 7 mixing boundary layer height (blue) derived from ERAI reanalysis and orography (black
 8 shading). (d) Tropospheric NO₂ distribution derived from GOME-2 measurements.
 9 Stratospheric ozone reaching (e) the LMT and (f) the atmospheric layer at 3-6 km of altitude,

- 1 | according to CHASER model simulations on 3 May 2009 at 10h00 JLT. Magenta rectangles
- 2 | show the locations of the air masses tracked during the pollution event in early May 2009.

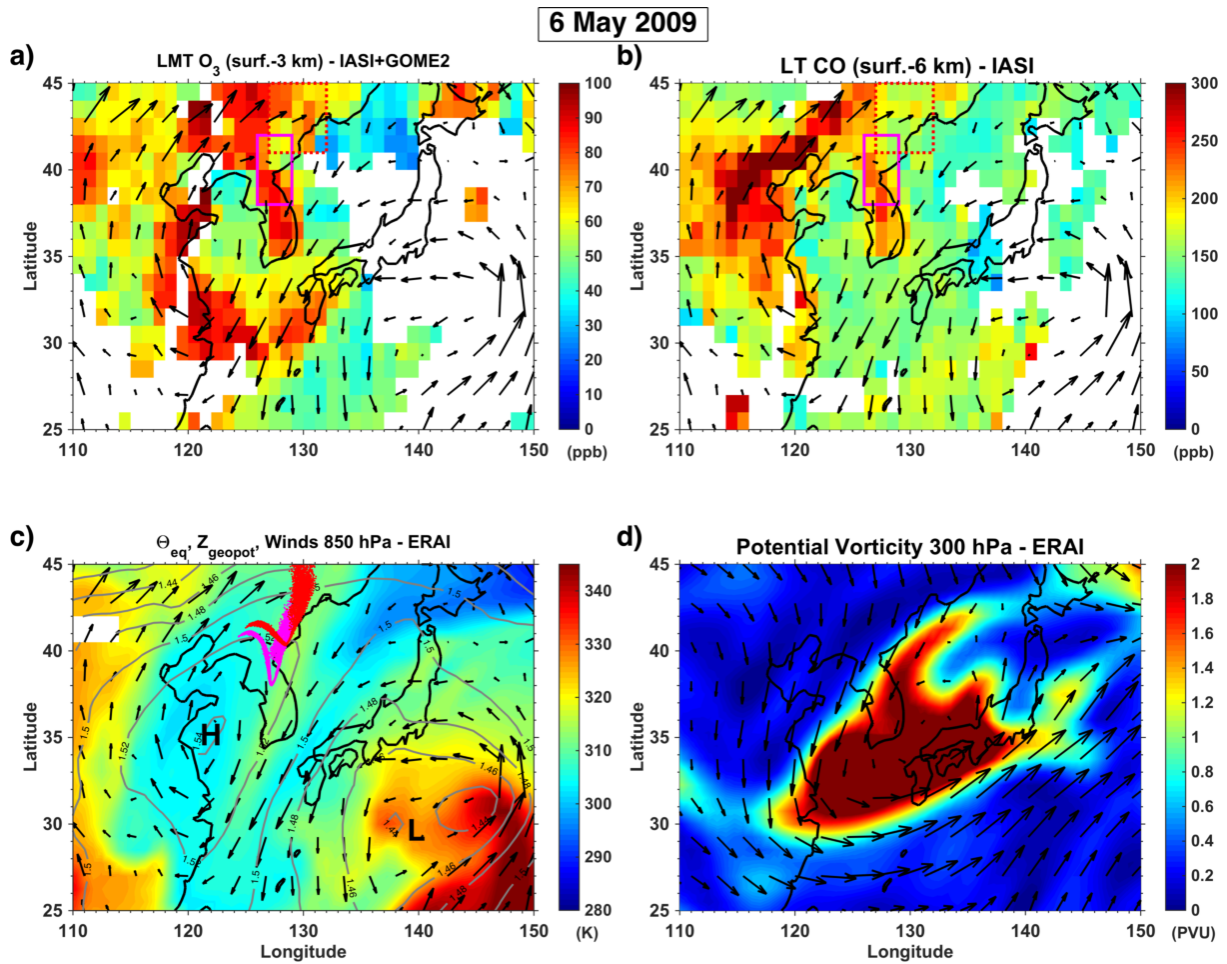


- 3
- 4 | **Figures 7.** Idem as Fig. 5 but for 4 May 2009.

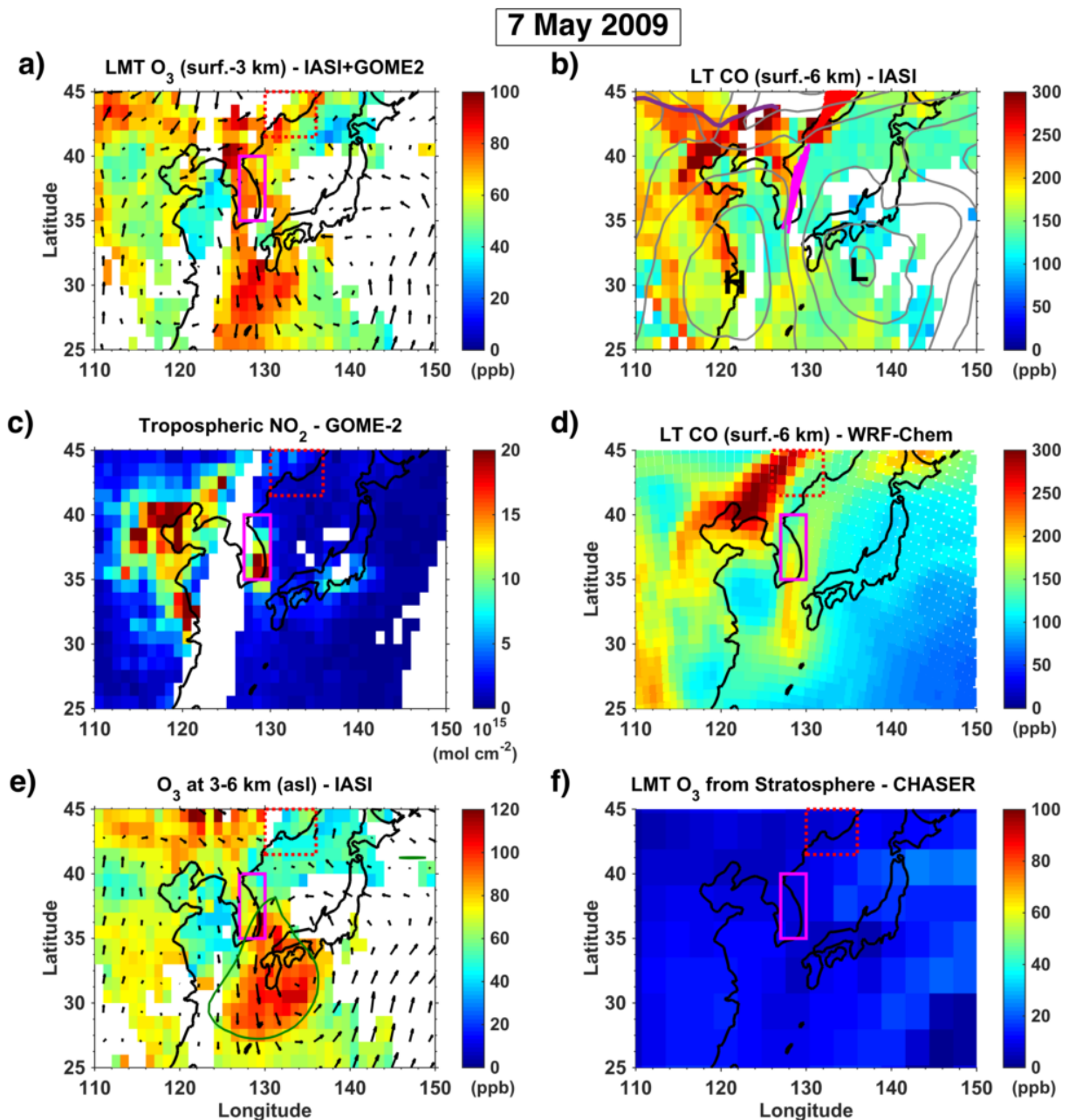


1

2 **Figure 8.** Idem as Fig. 7 but for 5 May 2009

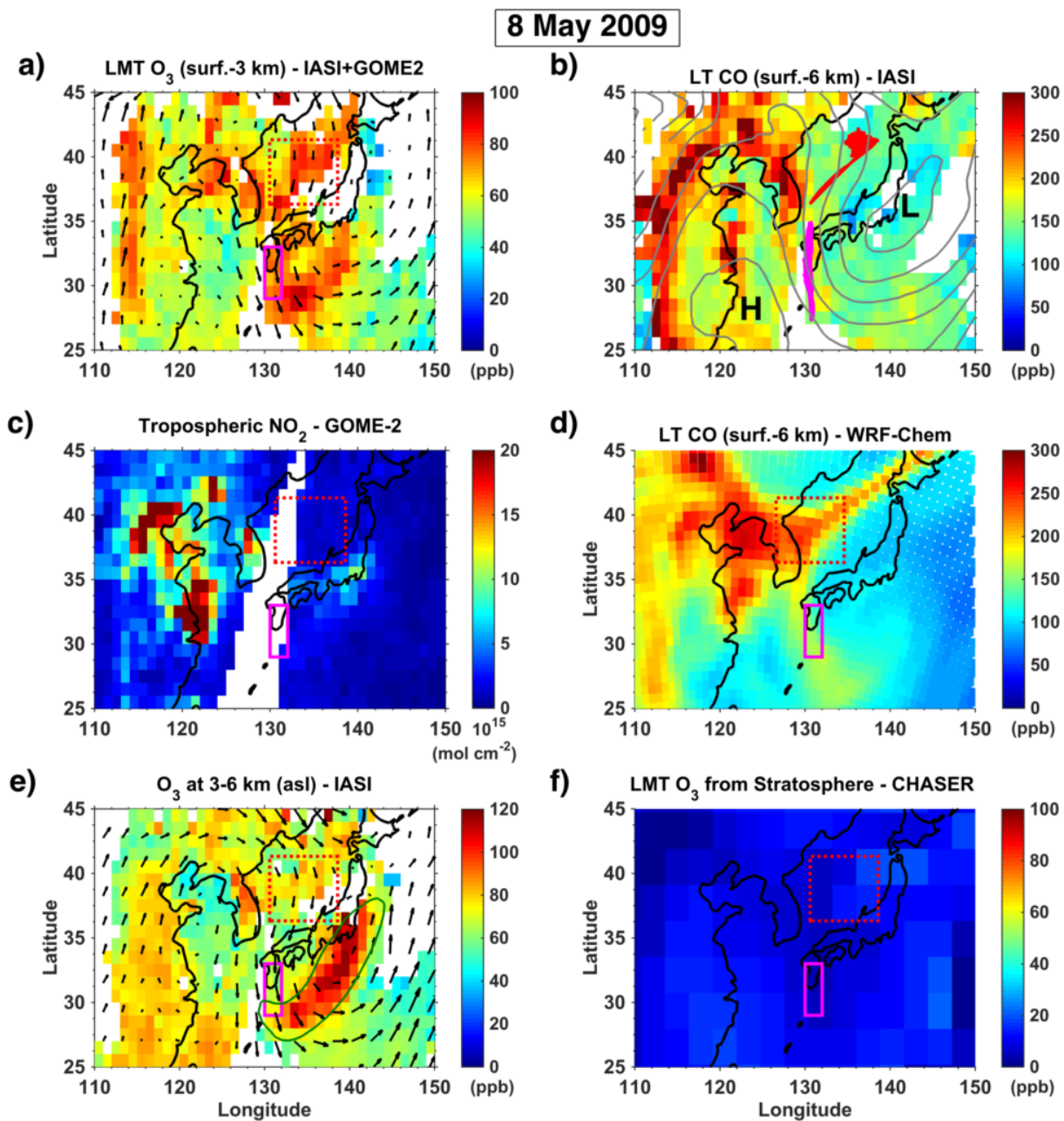


1
 2 **Figure 9.** Idem as Fig. 7 but for 6 May 2009. In panels (a-b), magenta and dotted red squares
 3 show the main location of the southern and northern pollution filaments, respectively. Dots in
 4 panel (c) indicate the location of the air parcels tracked with the HYSPLIT dispersion model,
 5 in magenta/red (southern/northern pollution plumes).

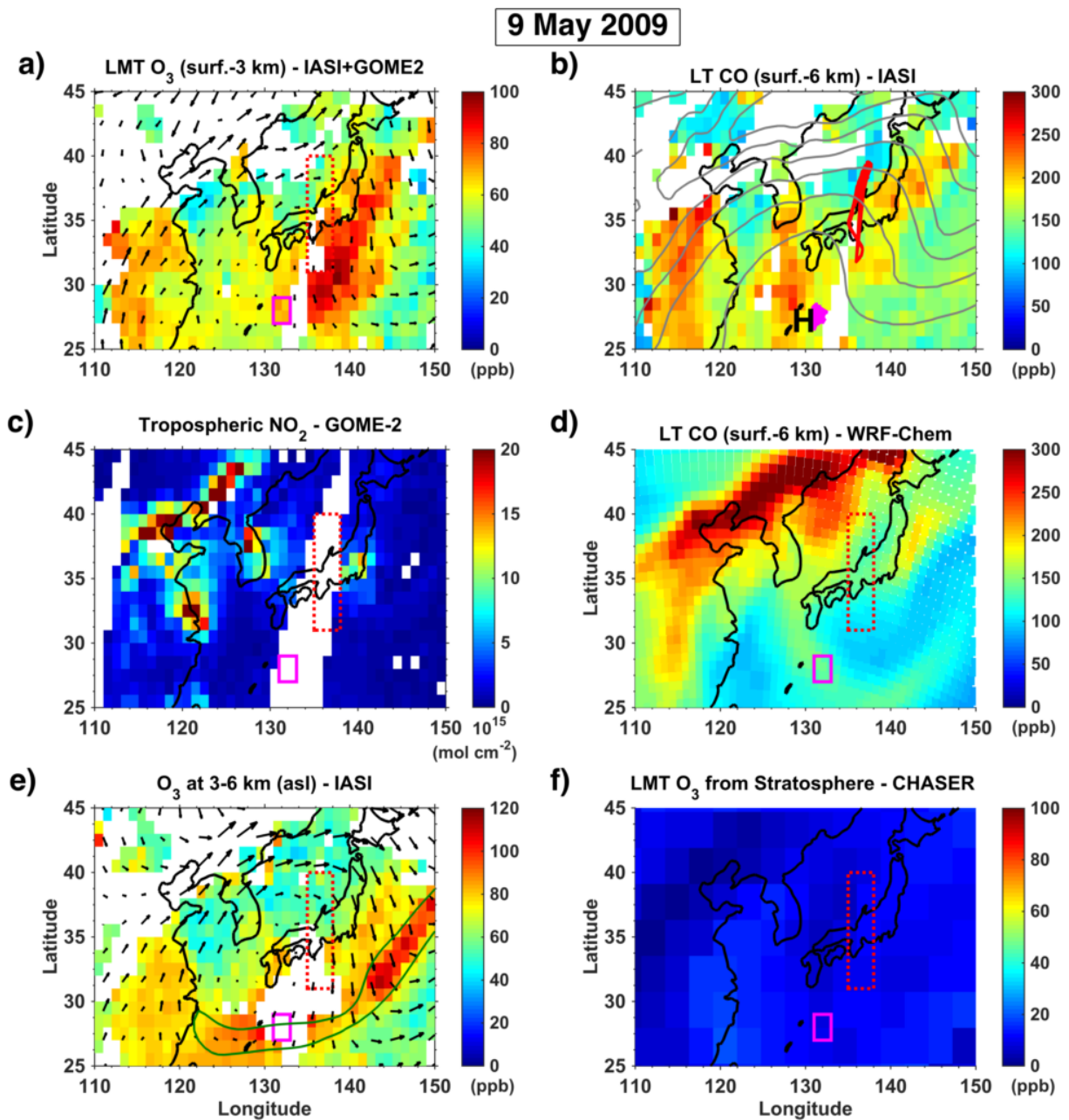


1
 2 **Figures 10.** Idem as Fig. 7 for panels (a) and (b) but for 7 May 2009. (c) Tropospheric NO₂
 3 distribution derived from GOME-2 measurements. (d) LT CO distribution according to WRF-
 4 Chem model. (e) Tropospheric ozone from 3 to 6 km of altitude derived from IASI, winds at
 5 700 hPa and potential vorticity at 300 hPa contours (2 PVU in green) from ERAI. (f)
 6 Stratospheric ozone reaching the LMT according to CHASER model simulations. Panel (b)
 7 also shows geopotential heights at 850 hPa from ERAI (grey contours every 200 m) and the
 8 location for 7 May 2009 of the polluted tracked air masses derived from HYSPLIT (magenta
 9 and red dots for the southern and northern pollution filaments). For WRF-Chem (panel d), the
 10 northern pollution plume (dotted red square) is shifted 4° to the west, in order to account for

1 the difference in its location between the model and satellite observations. The location of a
 2 cold front is shown by a violet curve in panel (b).

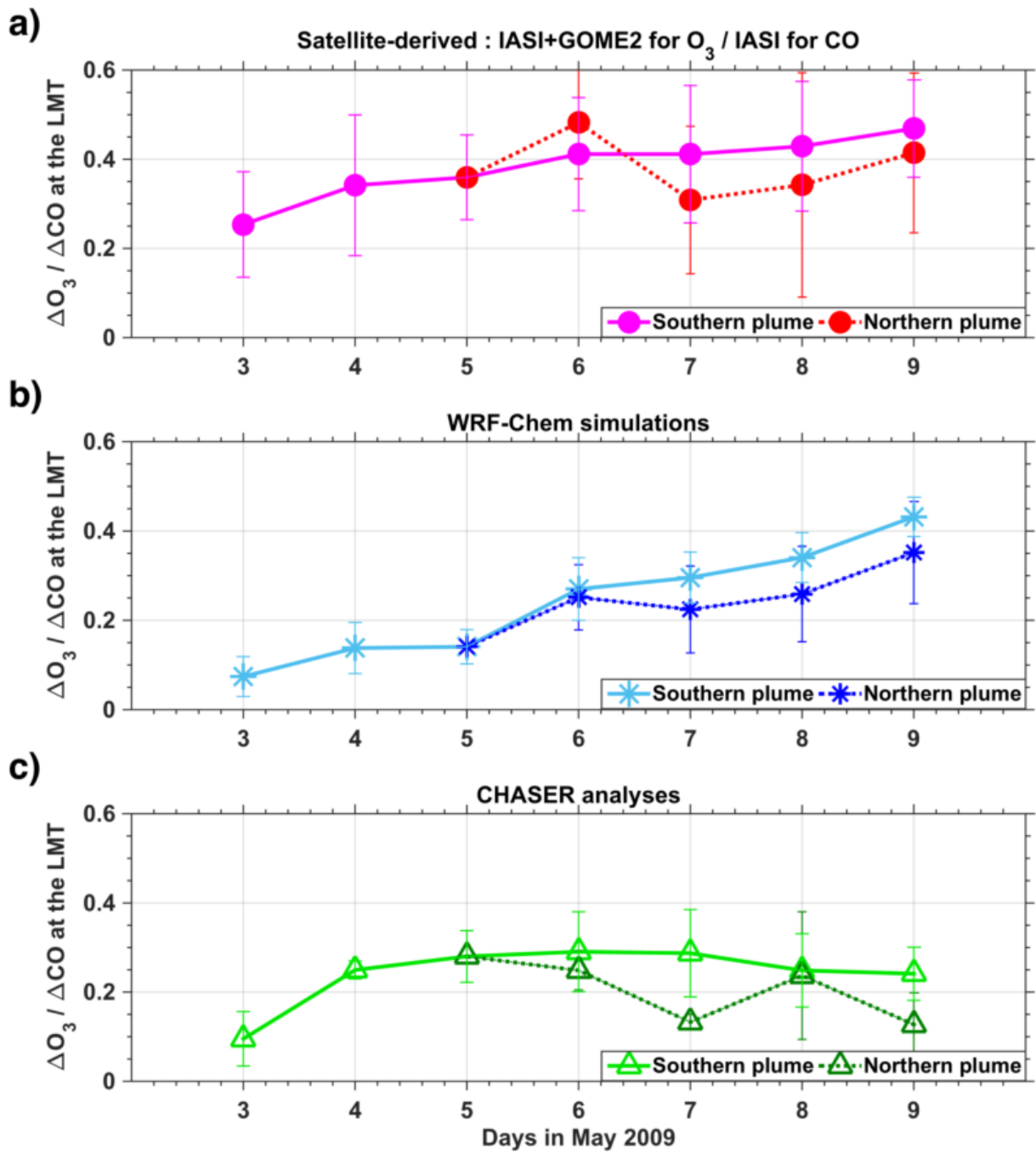


3
 4 **Figure 11.** Idem as Fig. 10 but for 8 May 2009.

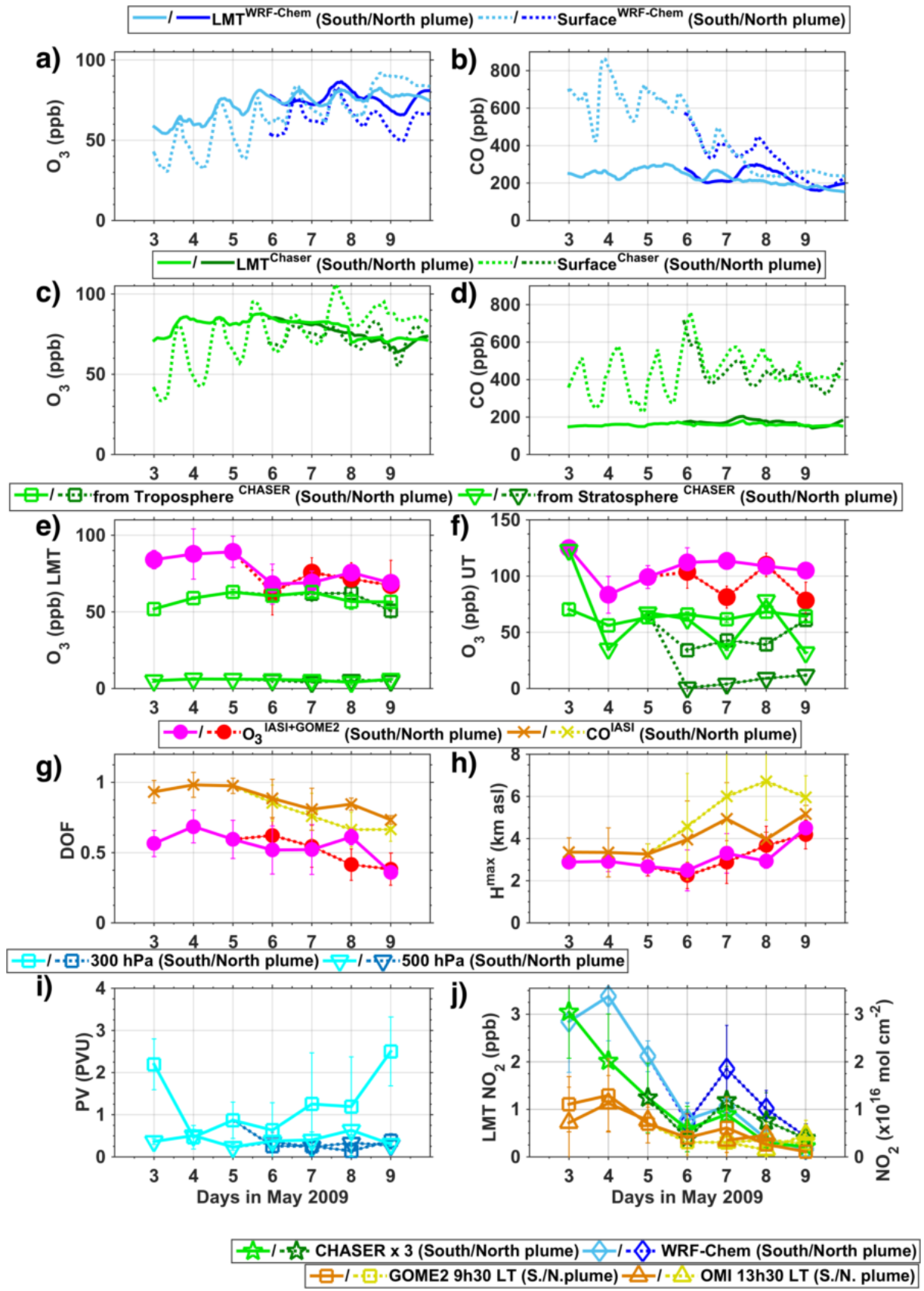


1

2 **Figures 12.** Idem as Fig. 10 but for 9 May 2009. Here, no shift is considered for the location
 3 of northern pollution plume (dotted red square) in WRF-Chem simulations, with respect to
 4 that of satellite observations.



1
 2 **Figure 13.** Lagrangian evolution of ozone enhancement along transport described by the ratio
 3 $\Delta O_3/\Delta CO$ at the LMT for the two pollution plumes tracked across East Asia from 3 to 9 May
 4 2009, derived from (a) IASI+GOME2 O₃ and IASI CO satellite retrievals, (b) WRF-Chem
 5 simulations and (c) CHASER analyses. Ratios of $\Delta O_3/\Delta CO$ for the southern (northern)
 6 pollution plumes are plotted in magenta (dotted red), light blue (dotted blue) and light green
 7 (dotted green) in respectively panels (a) to (c). Curves show mean and standard deviations
 8 (+/- vertical bars) of $\Delta O_3/\Delta CO$ over the areas depicted by rectangles in Figs. 5 and 7-11 for
 9 each of the days of the pollution outbreak.



1

1 **Figure 14.** Lagrangian evolution at the location of the pollution plumes across East Asia
2 (rectangles in Figs. 5 and 7-11) on 3-9 May 2009 for the following variables: **(a,c)** O₃ and
3 **(b,d)** CO mixing ratios at the LMT (plain lines) and the surface (dotted lines) from WRF-
4 Chem (respectively in panels a and b) and CHASER analysis (panels c and d), for the
5 southern (lighter colours) and northern (darker colours) pollution plumes. Ozone burden at the
6 **(e)** LMT and the **(f)** Upper Troposphere (UT), observed by IASI+GOME2 (magenta/red for
7 the southern/northern plumes) and simulated by CHASER for air masses originating from the
8 Troposphere (squares) and Stratosphere (triangles). Satellite retrievals sensitivity in term of
9 **(g)** degrees of freedom and **(h)** heights of maximum sensitivity at the LMT and LT for
10 respectively IASI+GOME2 and IASI. In panel **(g)**, DOFs for IASI+GOME2 are multiplied by
11 a factor 2 for visual clarity. **(i)** Potential vorticity at 300 (squares) and 500 hPa (triangles)
12 from ERAI reanalysis. **(j)** NO₂ concentrations observed as total columns by GOME-2 at 9h30
13 JLT (squares) and OMI at 13h30 JLT (triangles) and derived from WRF-Chem (ovals) and
14 CHASER analyses (stars, multiplied by a factor 3 for visual clarity) at the LMT at 10h00 JLT.
15 In panels **(e-j)**, curves with lighter and darker colours but the same marker correspond to
16 respectively the southern and northern pollution plumes. We show mean values and standard
17 deviations (+/- vertical bars) over the areas depicted by rectangles in Figs. 5 and 7-11 for each
18 of the days of the pollution outbreak.