

1 **Springtime daily variations of lower tropospheric ozone**
2 **over East Asia: role of cyclonic activity and pollution as**
3 **observed from space with IASI**

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17
18 **Abstract**

19 We use satellite observations from IASI (Infrared Atmospheric Sounding Interferometer) on
20 board the MetOp-A satellite to evaluate the springtime daily variations in lower tropospheric
21 ozone over East Asia. The availability of semi-independent columns of ozone from the
22 surface up to 12 km simultaneously with CO columns provides a powerful observational
23 dataset to diagnose the processes controlling tropospheric ozone enhancement at synoptic
24 scales. By combining IASI observations with meteorological reanalyses from ERA-Interim,
25 we elaborate an analysis method based only on IASI ozone and CO observations to identify
26 the respective roles of the stratospheric source and the photochemical source on ozone
27 distribution and variations over East Asia. The succession of low- and high-pressure systems
28 drives the day-to-day variations in lower tropospheric ozone. A case study analysis of one

1 frontal system and one cut-off low system in May 2008 shows that reversible subsiding and
2 ascending ozone transfers in the upper troposphere lower stratosphere (UTLS) region due to
3 the tropopause perturbations occurring in the vicinity of low-pressure systems impact free and
4 lower tropospheric ozone over large regions, especially north of 40°N, and largely explain the
5 ozone enhancement observed with IASI for these latitudes. Irreversible stratosphere-
6 troposphere exchanges of ozone-rich air masses occur more locally in the southern and south-
7 eastern flanks of the trough. The contribution to the lower tropospheric ozone column is
8 difficult to dissociate from the tropopause perturbations generated by weather systems. For
9 regions south of 40°N, a significant correlation has been found between lower tropospheric
10 ozone and carbon monoxide (CO) observations from IASI, especially over the North China
11 Plain (NCP). Considering carbon monoxide observations as a pollutant tracer, the O₃-CO
12 correlation indicates that the photochemical production of ozone from primary pollutants
13 emitted over such large polluted regions significantly contributes to the ozone enhancements
14 observed in the lower troposphere via IASI. When low-pressure systems circulate over the
15 NCP, stratospheric and pollution sources play a concomitant role in the ozone enhancement.
16 IASI's 3D observational capability allows the areas in which each source dominates to be
17 determined. Moreover, the studied cut-off low system has enough potential convective
18 capacity to uplift pollutants (ozone and CO) and to transport them to Japan. The increase of
19 the enhancement ratio of ozone to CO from 0.16 on 12 May over the North China Plain to
20 0.28 over the Sea of Japan on 14 May indicates photochemical processing during the plume
21 transport.

22

23 **1 Introduction**

24 In addition to being an important greenhouse gas (Stevenson et al., 2013), tropospheric ozone
25 (O₃) plays a central role in atmospheric chemistry and air quality, by controlling the oxidation
26 processes through the formation of hydroxyl radicals (OH) (Monks, 2005; Monks et al.,
27 2014). Ozone at high concentrations near the surface is a pernicious pollutant, harmful both to
28 human health and to vegetation (Seinfeld and Pandis, 1997, World Health Organization,
29 2013). Enhancements of ozone in the mid and lower troposphere result from photochemical
30 production from precursors (NO_x and hydrocarbons) and from stratosphere-troposphere
31 exchanges (STE) (Lelieveld and Dentener, 2000). The relative contributions made by these
32 sources depend on the season. It is well established that the peak activity of STE occurs

1 during winter and spring (Monks, 2000) whereas photochemical production is more active
2 during the summer period. The crucial role played by weather systems (cyclonic activity) in
3 determining tropospheric ozone variation has also been well established (e.g. Carmichael et
4 al., 1998; Cooper et al., 1998; Cooper et al., 2002a; Ding et al., 2009). These weather systems
5 are associated with tropopause perturbation, especially low tropopauses, and then with
6 subsiding and ascending ozone transfer in the upper troposphere – lower stratosphere (UTLS)
7 region. In addition, irreversible transfers of ozone can be expected, such as stratosphere-
8 troposphere exchanges that would take place preferentially on the western and southern flanks
9 of the trough (e.g. Ancellet et al., 1994; Holton et al., 1995; Liu et al., 2013), and downward
10 transport from the UTLS to the lower troposphere (e.g. Cooper et al., 2002a). Conceptual
11 models have been proposed to describe airstreams related to traveling low-pressure systems at
12 the midlatitudes (e.g. Cooper et al., 2002b). Two main mechanisms are responsible for part of
13 the ozone temporal and spatial variations observed in the troposphere. The dry airstream (DA)
14 occurring behind cold fronts is responsible for a strong downward transport of ozone from the
15 ULTS down to the middle troposphere. It is often linked to tropopause folding. This
16 downward transport can affect ozone concentrations down to the surface, especially at high
17 altitude sites (e.g. Carmichael et al., 1998; Schuepbach et al., 1999; Dempsey, 2014). In
18 contrast, air masses and then pollutants can be uplifted from the surface to the free
19 troposphere by different processes such as deep convection, orographic lifting and frontal
20 lifting (e.g. Bethan et al., 1998; Hannan et al., 2003; Miyazaki et al., 2003; Cooper et al.,
21 2004, Ding et al., 2009; Foret et al., 2014; Ding et al., 2015, and references therein). One part
22 of these processes, the warm conveyor belts (WCBs) associated with frontal activity and
23 lifting have been studied mainly in the scope of their role in the long-range transport of
24 pollutants, because they lift pollutants to levels where horizontal transport is more efficient.
25 Several studies focusing on the trans-Pacific transport of pollutants from East Asia towards
26 the United States have shown the importance of the frontal systems in this transport process
27 during springtime using both model simulations (e.g. Bey et al., 2001; Liu et al., 2003; Mari
28 et al., 2004; Lin et al., 2010) and dedicated field campaigns (e.g. Jaffe et al., 1999; Cooper et
29 al., 2004; Liang et al., 2004; Oshima et al., 2004). Very recently, Ding et al. (2015) have
30 shown that the topography of East Asia, as well as inducing orographic lifting, assists frontal
31 lifting and facilitates convection, thereby amplifying the possibility of pollutant uplifting.

32 In recent decades, East Asia and in particular China has experienced rapid economic growth.
33 The related increasing anthropogenic emissions of pollutants (Richter et al., 2005; Lin et al.,

1 2013) lead to regional ozone concentrations amongst the highest in the world (e.g. Chan and
2 Yao, 2008; Zhao et al., 2009; Lelieveld and Dentener, 2000; Wang et al., 2012; Safieddine et
3 al., 2013). Due to the rapidly changing emissions in China, the respective contribution made
4 by anthropogenic and natural perturbations to tropospheric ozone in China and its variability
5 constitutes a crucial issue to be documented and better understood. Seasonal variations in
6 ozone levels in East Asia and especially the role of the summer Asian monsoon leading to a
7 summer minimum have been extensively studied from model simulations, in situ and satellite
8 observations (e.g. Mauzerall et al., 2000; Tanimoto et al., 2005; Yamaji et al., 2006; Li et al.,
9 2007; Ding et al., 2008; Dufour et al., 2010). However, at the synoptic scale, the direct impact
10 of weather systems on tropospheric ozone distribution above China and its daily variations
11 has been less extensively considered or, if so, mainly in the scope of the long-range transport
12 of pollutants and export to the Pacific Ocean. A recent study investigates the dynamic and
13 chemical features induced in the upper troposphere by cut-off lows over northeast China
14 using limb and nadir satellite sounders (Liu et al., 2013).

15 The progress made in satellite observations of tropospheric ozone during the last decade (e.g.
16 Worden et al., 2007; Eremenko et al., 2008; Liu et al., 2010, Nakatani et al., 2012) offers a
17 new opportunity to evaluate ozone distribution and its daily variation including the role of
18 transport at the synoptic scale (e.g. Doche et al., 2014). The satellite provides an
19 unprecedented spatial coverage that allows new insight into how synoptic processes impact
20 ozone distributions. The first satellite measurements of tropospheric ozone were obtained
21 using ultraviolet-visible (UV) sounders (e.g. Fishmann et al., 2003; Liu et al., 2007). Later on,
22 the development of thermal infrared nadir sounders allowed accurate measurements of partial
23 tropospheric ozone columns to be obtained (Coheur et al., 2005; Worden et al., 2007; Dufour
24 et al., 2012; Safieddine et al., 2013). Using GOME and OMI UV sounders, Nakatani et al.
25 (2012) show a persistent belt of enhanced tropospheric columns of ozone at mid-latitudes
26 over East Asia throughout the year, partly attributed to stratospheric intrusion near the
27 subtropical jet. The tropospheric contribution to the enhanced ozone column has been
28 assessed using model simulations. Nakatani et al. (2012) underlined the difficulty in
29 differentiating the stratospheric and tropospheric origins of ozone in the tropospheric columns
30 observed by satellite. This difficulty has already been stated by de Laat et al. (2005).
31 However, it has been demonstrated that thermal infrared sounders like IASI on board MetOp
32 (Clerbaux et al., 2009) allow the retrieval of semi-independent partial columns of ozone
33 within the troposphere (Eremenko et al., 2008; Dufour et al., 2010; Dufour et al., 2012;

1 Safieddine et al., 2013; Barret et al., 2011). Dufour et al. (2010) show the ability of IASI to
2 provide independent information on the seasonal variation in lower and upper tropospheric
3 ozone over East Asia. Over shorter-term periods of the order of several days, the retrieved
4 ozone profile with IASI allows the identification of the origin of the observed tropospheric
5 ozone in specific cases. Very recently, Hayashida et al. (2015) show ozone enhancement in
6 the lower troposphere over East Asia using the OMI space-borne ultraviolet spectrometer.
7 They attribute the enhancement mainly to emissions of ozone precursors from open crop
8 residue burning after the winter wheat harvest.

9 In this paper, we use the IASI observation of lower tropospheric ozone to investigate the
10 influence of synoptic scale weather systems on the distribution of ozone over East Asia. In a
11 previous study, Dufour et al. (2010) show that IASI lower tropospheric ozone columns reach
12 a maximum in late spring and early summer (May, June) in Beijing, Shanghai and Hong
13 Kong. We then decided to focus our study on late spring (May), period during which high
14 ozone concentrations and frequent frontal activities occur over East Asia. We focus on May
15 2008, as this was the first period available with the new version of the IASI ozone product
16 used for this study. Two case studies associated with travelling low-pressure systems and
17 presenting enhanced ozone in the lower troposphere are analyzed. The first case is used to
18 elaborate the analysis method based on IASI observations of ozone (O_3) and carbon monoxide
19 (CO). We demonstrate that semi-independent ozone columns between the surface and 12 km
20 from IASI associated with simultaneous CO measurements provide a powerful observational
21 dataset to identify, at least partly, the stratospheric and anthropogenic origin of lower free
22 tropospheric ozone. The contributions made by descending air from the UTLS in the vicinity
23 of the weather systems and by the photochemical production of ozone to the enhanced lower
24 tropospheric ozone columns are then investigated for the two case studies.

25 The paper is structured as follow. In Section 2, the different satellite and meteorological
26 datasets are described. As a new version of the IASI ozone product is used for this study, we
27 provide a summary of the validation of the product with a specific focus on East Asia in
28 Section 3. The analysis method based on ozone and CO columns is detailed in Section 4.
29 Section 5 presents the consequences of a cut-off low travelling over a highly polluted region
30 (North China Plain) in terms of ozone vertical distribution and pollutant transport. A general
31 discussion is given in Section 6 as well as a conclusion in Section 7.

32

1 **2 Datasets description**

2 **2.1 The IASI instrument**

3 The IASI (Infrared Atmospheric Sounding Interferometer) (Clerbaux et al., 2009) instrument,
4 on board the MetOp-A platform since October 2006, is a nadir-viewing Fourier transform
5 spectrometer. It operates in the thermal infrared between 645 and 2760 cm^{-1} with an apodized
6 resolution of 0.5 cm^{-1} . The field of view of the instrument is composed of a 2×2 matrix of
7 pixels with a diameter at nadir of 12 km each. IASI scans the atmosphere with a swath width
8 of 2200 km and crosses the equator at two fixed local solar times 9:30 am (descending mode)
9 and 9:30 pm (ascending mode), allowing the monitoring of atmospheric composition twice a
10 day at any location. The large spectral coverage, high radiometric sensitivity and accuracy,
11 and rather high spectral resolution of the instrument allow this instrument to measure the
12 global distribution of several important atmospheric species (eg. Boynard et al., 2009; George
13 et al., 2009; Clarisse et al., 2011).

14 **2.2 Lower tropospheric ozone from IASI**

15 The IASI ozone profiles and partial columns considered in this paper have been retrieved
16 using the method described in Eremenko et al. (2008). The retrieval is performed using the
17 radiative transfer model KOPRA (Karlsruhe Optimised and Precise Radiative transfer
18 Algorithm) and its inversion module KOPRAFIT (Stiller et al., 2000; Höpfner et al., 2001),
19 both adapted to the nadir-viewing geometry. A constrained least squares fit method with an
20 analytical altitude-dependent regularization is used (Kulawik et al., 2006). The applied
21 regularization method is detailed in Eremenko et al. (2008). To summarize, the regularization
22 matrix is a combination of first order Tikhonov constraints (Tikhonov, 1963) with altitude-
23 dependent coefficients. The coefficients are optimized both to maximize the degrees of
24 freedom (DOF) of the retrieval and to minimize the total error on the retrieved profile.
25 Compared to previous studies using this algorithm (Eremenko et al., 2008; Dufour et al.,
26 2010, 2012), several changes have been made. The emissivity of the surface is now taken into
27 account based on a global monthly IASI-derived climatology (Zhou et al., 2011) allowing a
28 better retrieval above arid regions. Different a priori and constraints are used depending on
29 the tropopause height. This new scheme was introduced to reduce possible compensation
30 effects during the retrieval procedure. An automatic detection of the tropopause height
31 (calculated from the temperature profile retrieved from IASI using the definition based on the

1 lapse rate criterion (WMO, 1957)) has been introduced to discriminate between polar,
2 midlatitudes, and tropical situations. If the tropopause is lower than 10 km, the polar
3 constraint and a priori profile are used. If the tropopause is between 10 and 14 km, the
4 midlatitude constraint and a priori profile are used. If the tropopause is higher than 14 km, the
5 tropical constraint and a priori are used. The midlatitude and tropical regularization matrices
6 are those already used in Eremenko et al. (2008) and Dufour et al. (2010, 2012) respectively.
7 The polar constraint has been specifically developed following the same method as in
8 Eremenko et al. (2008). The a priori profiles are compiled from the ozonesonde climatology
9 of McPeters et al. (2007). The midlatitude a priori profile is set to the climatological profile of
10 the 30-60°N latitude band for summer. The tropical a priori profile is set to the climatological
11 profile of the 10-30°N latitude band over the year. The polar profile is set to the
12 climatological profile of the 60-90°N latitude band for summer. As the version of the ozone
13 product used in this study differs significantly from the version extensively validated in
14 Dufour et al. (2012), a new validation against ozonesondes has been conducted and the results
15 are presented in Section 3. The modifications of the algorithm do not influence the vertical
16 sensitivity of IASI. As shown in Dufour et al. (2010, 2012), two semi-independent partial
17 columns of ozone between the surface and 12 km can be considered: the lower tropospheric
18 column integrating the ozone profile from the surface to 6 km altitude – above sea level (asl)
19 – and the upper tropospheric column integrating the ozone profile from 6 to 12 km altitude.
20 Note that the latter column can include stratospheric air masses depending on the tropopause
21 height. The averaging kernels give information on the vertical sensitivity and resolution of the
22 retrieval. The lower tropospheric column shows a maximum sensitivity typically between 3
23 and 4 km with a limited sensitivity to the surface (Dufour et al., 2012). This implies that the
24 ozone concentration profile in the lower troposphere is preferentially incremented at these
25 altitudes during the retrieval process, independently if the true ozone profile is perturbed at
26 other altitudes, especially at the surface. Moreover, it is worth noting that the partial columns
27 are only semi-independent which means that they may include partial information from
28 altitudes outside their altitude range. For example, the lower tropospheric column includes
29 information from altitudes higher than its upper limit (6 km). In order to estimate the fraction
30 of contamination of the lower tropospheric column by higher altitudes, we calculated the ratio
31 between the integral of the averaging kernel of the lower tropospheric column from 6 km to
32 60 km and the integral from the surface to 60 km. Higher atmospheric layers contribute to
33 about 20 to 30 % of the lower tropospheric column in the midlatitude air masses (not shown).

1 Note that only the morning overpasses of IASI are considered for this study in order to remain
2 in thermal conditions with a better sensitivity to the lower troposphere.

3 **2.3 Carbon monoxide from IASI**

4 The CO data used here are retrieved from the IASI spectra within the 2143-2181.25 cm^{-1}
5 spectral range using the FORLI-CO retrieval code from the Université Libre de Bruxelles
6 (ULB). FORLI-CO retrievals give CO concentration profiles using the optimal estimation
7 method (Rodgers, 2000) and a single a priori profile. More details are given in Hurtmans et al.
8 (2012). The IASI FORLI-CO product used in this study is the total column, publicly available
9 from the Ether website (<http://www.pole-ether.fr>). Note that only half of the pixels are
10 available for the year 2008. This explains the difference in measurement density between O_3
11 and CO observations in the different figures. Carbon monoxide is often used as an indicator of
12 biomass burning and anthropogenic pollution (e.g. Edwards et al., 2004; McMillan et al.,
13 2010). In this study, we use the IASI CO columns as an anthropogenic pollution tracer.

14 **2.4 Meteorological dataset**

15 Meteorological data from the ECMWF ERA-Interim reanalysis are used in our analyses. The
16 reanalysis is based on a 4D-Var assimilation system with a 12-hour analysis window. The
17 spatial resolution of the data set is approximately 80 km on 60 vertical levels from the surface
18 up to 0.1 hPa (Dee et al., 2011). In our analyses, the meteorological parameters are taken at
19 0:00 UTC, corresponding roughly to the morning overpass time of IASI. The main variables
20 considered in this study are the geopotential height, the potential vorticity (PV), and the
21 horizontal wind field (u and v components), as well as the equivalent potential temperature,
22 the vertical velocity and the convective available potential energy. The geopotential height
23 associated with horizontal wind at 850 hPa give a proxy for describing the weather situation
24 and horizontal transport in the lower troposphere, whereas the same parameters at 300 hPa
25 describe the situation in the UTLS. We also calculate the equivalent potential temperature at
26 850 hPa and 300 hPa from temperature, relative humidity and specific humidity fields
27 (Bolton, 1980) as an indicator of air masses origin (Holton, 2004). Potential vorticity (PV) is
28 often used as a tracer of tropopause height and of air masses origin (e.g. Bethan et al., 1996).
29 PV values between 1 and 1.6 PVU are representative of the upper troposphere whereas PV
30 values larger than 1.6 PVU are indicators of air mass origin above the dynamical tropopause.
31 In this study, we consider mainly PV averaged at between 300 and 500 hPa with a 50 hPa

1 interval as we are above all interested in the impact of stratospheric air masses on the free
2 troposphere. In order to investigate the ascending motion of air masses, especially from the
3 boundary layer towards the free troposphere within weather systems, we examine the vertical
4 velocity at different pressure levels as well as the convective available potential energy
5 (CAPE), which informs on the capability of the low-pressure system to vertically transport air
6 masses by convection.

7

8 **3 Validation of IASI lower tropospheric ozone**

9 Significant changes in the ozone retrieval procedure compared to the validation exercise
10 reported in Dufour et al. (2012) have been made as described in Section 2.2. A new validation
11 exercise was done to evaluate the new version of the ozone product. We use a database of
12 ozonesonde measurements from 2007 to 2012 including 27 stations in the midlatitudinal band
13 (30-60°) in both hemispheres and 16 stations in the tropical band (30°S-30°N). Most of the
14 ozonesonde measurements are from the WOUDC (<http://woudc.org/>) and SHADOZ
15 (<http://croc.gsfc.nasa.gov/shadoz/>) databases, except for Aquila and Beijing. A list of stations
16 and related information is provided in Table 1. The coincidence criteria used for the
17 validation are 1° around the station, a time difference smaller than 6 hours and a minimum of
18 10 clear-sky pixels matching these criteria. The results of the comparison between IASI ozone
19 retrievals and ozonesonde measurements are summarized in Table 2. We focus on the lower
20 troposphere and then no correction factor has been applied on ozonesonde measurements. The
21 results for other partial columns are not significantly different compared to the previous
22 version of the product, extensively discussed by Dufour et al. (2012). The bias for the lower
23 tropospheric column (surface to 6 km asl) is small -0.6 DU (-2.8%) and comparable to the
24 bias estimated at the midlatitudes with the previous version of the product (Dufour et al.,
25 2012). The estimated error is about 2.8 DU (14%) with a correlation coefficient of 0.70. Table
26 2 also summarizes the results for East Asian ozonesonde stations only (Beijing, Hong Kong,
27 Sapporo and Tateno). A significant bias of 2.2 DU (9.5%) with IASI underestimating ozone
28 partial columns is determined. The bias is similar for Beijing, Hong Kong, and Tateno (-2.6
29 DU) and different for Sapporo (+0.8 DU). Most of the ozonesonde measurements are
30 performed in the early afternoon. The ozone build-up is then maximal in polluted urban or
31 suburban sites like Beijing, Hong Kong, and Tateno. IASI observations are performed in the
32 morning, about 5 hours earlier on average. The time difference between IASI and ozonesonde

1 observations in polluted suburban sites may partly explain the larger bias in this case. Indeed,
2 the bias for the Sapporo region, where the diurnal cycle of ozone is limited, is reduced.
3 However, the small number of coincidences does not allow any firm conclusion to be reached
4 on the origin of the observed bias over East Asia.

5

6 **4 Case study of 4-6 May 2008: on the use of IASI O₃ and CO to diagnose the** 7 **processes influencing the ozone distribution affected by weather systems**

8 An episode of high ozone is observed in the lower troposphere with IASI in North East Asia
9 from 4 to 6 May 2008. This episode is associated with a low-pressure system travelling from
10 Mongolia through North China to the extreme north of Japan. In this section, we investigate
11 how to use the ozone partial columns and profiles and the CO total columns from IASI to
12 diagnose which processes contribute to the ozone enhancement.

13 **4.1 Low-pressure system and associated IASI ozone distribution**

14 Figure 1 describes the meteorological situation of this episode of high ozone. A large cold
15 front extending from Mongolia to South China on 4 May 2008, from North China to the
16 Southern Japanese Islands on 5 May 2008, and moving eastward from Japan on 6 May 2008
17 characterizes the low-pressure system (Figs. 1a-c). The regions behind the frontal area and
18 north to the polar jet, situated between 35°N and 40°N on these dates (Figs. 1d-f), are strongly
19 influenced by polar air masses with tropopause heights below 9 km (Figs. 1g-i). The 300-500
20 hPa mean PV values are larger than 1.6 PVU for the same regions, indicating that the upper
21 troposphere is under the influence of lower stratospheric air masses (Figs. 2j-l). The spatial
22 correlation of low tropopauses and large PV values indicates that reversible subsiding ozone
23 transfer affects the upper troposphere in this case. We then expect an ozone enhancement in
24 the upper troposphere for these regions and we will see in the following how IASI describes
25 this ozone enhancement induced by ozone subsidence. The analysis of the upper tropospheric
26 columns shows that IASI observes columns larger than 40 DU in the regions affected by low
27 tropopauses and large PV (Figs 2g-i). A step gradient between 30 and 40 DU is observed in
28 the upper tropospheric ozone distribution reflecting the step gradient in the PV distribution.
29 The very good spatial correlation of the high UT ozone structures with those of high PV leads
30 us to consider that the upper tropospheric columns of ozone retrieved from IASI can be used

1 as a proxy to determine the regions affected by subsiding ozone from the lower stratosphere.
2 The threshold of 40 DU seems to be relevant for this identification.

3 The question now is to determine to what extent IASI is able to inform about the low-pressure
4 system's influence on the lower tropospheric ozone distribution. The physical processes,
5 which may affect the lower tropospheric ozone distribution, are (i) the reversible ozone
6 subsidence associated with low tropopause heights, which induces an enhancement of ozone
7 in the upper and free troposphere, and then partly in the lower troposphere; (ii) irreversible
8 stratosphere-troposphere exchanges, which also lead to ozone enhancement. The first process
9 is expected to affect ozone distribution at a synoptic scale whereas the second process is more
10 localized. Figures 2a-c show that lower tropospheric ozone columns larger than 28 DU are
11 observed with IASI in the vicinity of the low with similar spatial patterns to the UT columns
12 and the PV distribution. The observed enhancement in the lower tropospheric column
13 (surface-6km) arises from

- 14 • the actual (reversible) transfer of ozone to the free troposphere;
- 15 • the definition of the LT columns by itself. The upper boundary of the column is fixed
16 at 6 km. When the tropopause is low (below 9 km), the LT column arithmetically
17 includes layers with upper tropospheric characteristics;
- 18 • the limited vertical resolution of the retrieval and the associated smoothing of the
19 vertical profile. As discussed in Section 2.2, the lower tropospheric column is then
20 partly contaminated by ozone outside the column altitude boundaries. This may
21 contribute to an overestimation of the lower tropospheric column. However, it is
22 difficult to estimate this overestimation in our case because no ozonesonde
23 observations were available along the path of the low.

24 We show with this case study that having the IASI UT and LT ozone columns allows us to
25 determine those regions affected by the subsiding transfer of ozone occurring behind the
26 frontal area and if the lower troposphere is affected. Now, we will examine if and how IASI
27 can be used to characterize irreversible stratosphere-troposphere exchanges (STE). The
28 analysis of the PV distribution at different pressure levels allows the identification of the
29 region in the vicinity of the low where STE occurs. In the case study from 4 to 6 May 2008,
30 we identify two regions with high PV values down to 600 or 500 hPa on the path of the low
31 (not shown): one on the east coast of Korea on 5 May ($\sim 39^\circ\text{N}$, 128°E) and one offshore to the
32 northeast of Tokyo ($\sim 39^\circ\text{N}$, 142.5°E). The STE is situated in the southeast flank of the low-

1 pressure system, behind and in the southern part of the cold front in the two cases. Figure 3
2 displays the longitudinal and latitudinal vertical section of ozone at 128°E and 39°N for 5
3 May (top) and at 142.5°E and 39°N for 6 May (bottom). On 5 May, strong stratospheric
4 intrusion of ozone is observed between 38°N and 39°N, and between 125°E and 130°E. The
5 vertical section at 128°E shows that the free and lower troposphere are still connected to the
6 polar UTLS reservoir that day. On 6 May, a stratospheric intrusion is observed between 36°N
7 and 38°N, and between 140°E and 143°E. The vertical section along 142°E shows that the
8 enhanced ozone in the lower troposphere (below 7 km) is partly disconnected from the polar
9 UTLS reservoir. Backtrajectories performed with the HYSPLIT trajectory model (Draxler and
10 Rolph; Rolph) show that the 3km-altitude air masses located in this area originate from
11 altitudes between 5 and 7 km the day before from North China and Inner Mongolia (Fig. S1).
12 The tropopause height was around 7-8 km on 5 May 2008 for these regions (Fig. 1h). This
13 means that the air masses reaching northeast of Tokyo at 3 km on 6 May have a UTLS origin
14 and transport ozone-rich air into the lower troposphere. Thus, we show that the downward
15 transport from the UTLS affects ozone concentrations in the lower troposphere for specific
16 regions on the southeastern flank of the weather system, whereas the perturbation of the
17 tropopause associated with this system influences upper and lower tropospheric ozone over
18 larger areas in the vicinity of the low.

19 **4.2 Influence of the high-pressure system on tropospheric ozone distribution** 20 **over the NCP**

21 On 5 May 2008, an anticyclone is forming over Central East China and the North China Plain.
22 The northwesterly winds reaching the NCP change progressively to southwesterly winds from
23 4 to 6 May with low winds and then a stagnant situation on 5 May (Figs. 1a-c). This,
24 associated with low cloud coverage and then increasing radiation, is a situation favorable to
25 the accumulation of primary pollutants over the NCP and to the photochemical production of
26 ozone due to both local emissions and regional transport of pollutants. The question here is
27 how IASI is able to describe this situation. The CO columns observed with IASI show
28 enhanced values over the NCP on 5 and 6 May (Figs. 2e and 2f). Considering CO as a
29 pollution tracer, enhanced IASI CO columns can be used to evaluate the build-up of
30 pollutants. Concomitantly, lower tropospheric ozone columns as large as 30 DU are observed
31 (Figs. 2b and 2c). A significant spatial correlation ($r=0.6$) is calculated between CO and lower
32 tropospheric ozone columns for a square region including the NCP (35-41°N, 114-122°E) on

1 5 May. In addition, the upper tropospheric ozone column does not show enhanced values over
2 the NCP for these 2 days (Figs. 2h and 2i). The analysis of the vertical section of ozone
3 distribution on 5 May shows that the large ozone concentrations in the Beijing region (Fig. 3,
4 $\sim 39^\circ\text{N}$ and $\sim 116^\circ\text{E}$) and across the NCP are retrieved below 6 km and are disconnected from
5 the UTLS region. The maximal values of ozone are retrieved between 2 and 3 km over
6 Beijing (Fig. 3) in agreement, considering the vertical sensitivity and resolution of IASI, with
7 in situ measurements, which frequently report high ozone concentrations at an altitude of 1.5-
8 2km above Beijing during April-May (Huang et al., 2014). This associated with the
9 correlation with CO suggests that the enhanced ozone observed with IASI is mainly due to the
10 photochemical transformation of primary pollutants emitted over the NCP. To evaluate the
11 degree of photochemical production of ozone, we calculate the equivalent or mean mixing
12 ratio corresponding to the CO and LT O₃ columns. This allows us to estimate a relative
13 enhancement ratio of O₃ to CO of 0.14 and 0.08 on 5 and 6 May respectively. These values
14 are in agreement with the typical values ranging between 0 and 0.3 reported over East Asia by
15 Tanimoto et al. (2008). The estimated enhancement ratio remains quite low suggesting an
16 early stage of ozone production.

17

18 **5 Case study of 11-16 May 2008: combined contributions of anthropogenic** 19 **and stratospheric sources over the NCP and pollution transport**

20 A second episode of high ozone is observed in the lower troposphere over the North China
21 Plain (NCP) from 11 to 16 May 2008. This episode is associated with a cut-off low-pressure
22 system forming on 11 May over Inner Mongolia and moving east on subsequent days. From
23 14 May, the meteorological regime changes over the NCP with warmer air masses settling
24 within an anticyclonic situation. In this section, we examine the influence of the
25 meteorological situation on the distribution of lower and upper tropospheric ozone with a
26 particular focus on the NCP. Figure 4 describes the meteorological situation for the entire
27 period. Figures 5 and 6 display the lower and upper tropospheric ozone columns and the total
28 CO columns observed with IASI.

29 **5.1 11-13 May: NCP under the direct influence of the cut-off low**

30 On 11 May 2008, a cut-off low is forming over Inner Mongolia (Fig. 4a). The cut-off low is
31 not yet completely dissociated from the polar reservoir. A band of upper tropospheric

1 columns larger than 40 DU is observed by IASI between 35°N and 45°N (Fig. 5g). As seen in
2 Section 4, it indicates that the region is under the influence of subsiding ozone. The lower
3 tropospheric ozone columns do not show a clear enhancement for the same latitude band. On
4 that day, subsiding ozone affects only moderately the lower tropospheric ozone.

5 On 12 May 2008, the cut-off low is well dissociated from the western current and its center
6 reaches the Bohai Sea (Fig. 4b). Upper tropospheric ozone columns larger than 45 DU are
7 retrieved all around the cut-off low. Lower tropospheric ozone columns larger than 32 DU are
8 observed especially in the southwestern part of the low, just above the NCP (Fig. 5b). The
9 analysis of the vertical section of ozone at 117°E (Fig. 7a) shows that the subsiding transfer of
10 ozone due to the tropopause perturbation strongly affects lower tropospheric ozone north of
11 33°N. At 32°N, the ozone enhancement observed in the lower troposphere is not connected to
12 the UTLS reservoir, suggesting a possible photochemical origin for this enhancement. IASI
13 CO columns are also enhanced in the NCP region and partly correlated with the enhanced
14 ozone columns (Fig. 5e). This indicates that pollution likely plays a concomitant role in the
15 ozone enhancement in that case.

16 On 13 May 2008, the centre of the cut-off low moves slightly to the East and reaches the
17 Yellow Sea (Fig. 4c). As for the previous day, large upper and lower tropospheric ozone
18 columns are observed with IASI in the vicinity of the low (Figs. 5c and 5i). The two columns
19 are slightly smaller than the day before over the NCP. The analysis of the vertical section of
20 ozone at 115°E (Fig. 7b) shows that the subsiding transfer of ozone due to the tropopause
21 perturbation is less effective than the previous day. Even if the lower tropospheric ozone
22 remains partly connected to the UTLS reservoir north of 33°N, secondary maxima are
23 observed at ~4 km of altitude, suggesting that an additional source of ozone may contribute to
24 the LT ozone enhancement. South of 33°N the ozone enhancement is clearly located in the
25 lower troposphere. The good spatial correlation of LT ozone enhancement and the strong CO
26 enhancement observed all over the NCP (Fig. 5f) confirms that pollution plays a concomitant
27 role in explaining the ozone distribution in the lower troposphere over the NCP for this day.

28 **5.2 14 May: transition from a cyclonic to an anticyclonic situation**

29 On 14 May 2008, the cut-off low shifts to the Sea of Japan (Fig. 4d). A large area including
30 North China, Korea, and reaching Japan shows upper tropospheric columns larger than 40 DU
31 (Fig. 6g), which indicates the region is under the influence of subsiding ozone. Within this

1 area, the largest LT ozone columns are observed in an area less extended and situated on the
2 southeastern flank of the low, mainly over the Sea of Japan (Fig. 6a). The lower troposphere
3 is then certainly under the influence of the UTLS.

4 Over China, an anticyclonic situation starts to develop south of the NCP inducing a change in
5 the wind regime and warmer conditions from 14 May (Fig. 4d). Enhanced CO columns and
6 lower tropospheric ozone columns are retrieved with IASI over the NCP (Figs. 6a and 6d)
7 with moderate UT ozone columns. The analysis of the vertical section of ozone
8 concentrations at 35°N shows that very large ozone concentrations are retrieved for the entire
9 free and upper troposphere in the eastern part of the section (Fig. 7c). This corresponds to the
10 region over the Sea of Japan under the direct influence of the cut-off low and then greatly
11 influenced by the UTLS. The situation is different over the NCP: ozone concentrations in the
12 upper troposphere are moderate and a distinct maximum in the lower troposphere is clearly
13 visible. This, associated with CO enhancement over the NCP in good spatial correlation with
14 LT ozone, indicates that the ozone enhancement observed with IASI over the NCP is of
15 anthropogenic origin and related to the photochemical production of ozone. On that day, the
16 estimated enhancement ratio of O₃ to CO is 0.11 in agreement with the enhancement ratio
17 calculated for the previous case study.

18 **5.3 15-16 May: NCP under anticyclonic influence**

19 On 15 and 16 May 2008, strong enhancements of CO and lower tropospheric ozone are
20 observed with IASI over the entire NCP (Figs. 6b-c and 6e-f). Both CO and O₃ increase
21 compared to the previous day. The anticyclone is firmly settled over China, leading to a
22 stagnant situation with low winds all over the NCP (Fig. 4e). This situation is favorable to the
23 accumulation of pollutant and then to the photochemical production of ozone. Figure 7d
24 shows the vertical section of ozone concentrations retrieved with IASI at 37°N. The ozone
25 enhancement is located below 4 km, especially between 115°E and 116°E, in agreement with
26 the findings of Section 4.2. This, with CO enhancement, indicates that the ozone enhancement
27 is due to photochemical production from pollutants emitted in the NCP. In this case of
28 stronger CO enhancement, the enhancement ratio of O₃ to CO (0.09 on 15 May and 0.06 on
29 16 May) decreases compared to the previous days.

1 **5.4 Evidence of transboundary transport within the cut-off low**

2 On 13 and 14 May, large CO and O₃ columns are retrieved from IASI over the Yellow Sea
3 and over the Sea of Japan on the southern flank of the cut-off low-pressure system (Figs. 5c
4 and 6a). Fairly strong westerly winds are present at 850 hPa in the same region, suggesting a
5 possible advection of air masses from the NCP towards Japan associated with the weather
6 system (Figs. 4c and 4d). In order to assess whether the weather system may have contributed
7 to transporting the pollutants (O₃ and CO), we perform backtrajectories on 13 May for an area
8 south to Korea (Fig. S2). The 3-km air masses originate from the boundary layer over NCP
9 on 11 May. They have been uplifted and transported at an altitude of between 3 and 4 km on
10 subsequent days (Fig. S2). In order to investigate if the pollutant uplifting on 11 May occurs
11 over a region more extended than those shown on Fig. S2, we examined two meteorological
12 variables that indicate possible ascending motion of air masses: the convective available
13 potential energy (CAPE) and the vertical velocity. Figure 8 shows that CAPE is significant on
14 the inside eastern flank of the cut-off low and that negative vertical velocities, i.e., ascending
15 winds, are present from the surface up to 300 hPa (Fig. 8 shows only the vertical velocity at
16 700 hPa as an example). In addition, backtrajectories performed on 11 May indicate that most
17 of the air masses between 38-40°N and 116-117°E at 3 km originate from the atmospheric
18 layers below 1 km and circulate over the NCP during the previous 24 hours (Fig. S3). This
19 evidences that pollutants (CO and O₃) have been uplifted from the boundary layer into the
20 free troposphere over NCP and then exported towards Japan by the cut-off low. This transport
21 pathway is relatively well known. Very recently, Ding et al. (2015) studied in detail the
22 uplifting and transport of CO in East Asia. They show that the vertical transport of
23 anthropogenic CO originating from the NCP is mainly carried out by frontal lifting associated
24 or not with WCB. They also pointed out the additional topography's role in the CO lifting
25 over the NCP.

26 To complete the study, we calculate the enhancement ratio of O₃ to CO over NCP on 12 May,
27 over the Yellow Sea and Korea (32-36°N, 122-130°E) on 13 May, and over the Sea of Japan
28 (30-38°N, 128-140°E) on 14 May. The ratios are respectively 0.16, 0.21 and 0.28. The
29 increase of the ratio indicates possible photochemical processing during the transport. Part of
30 the large lower tropospheric ozone is then due to the transport of ozone produced over the
31 NCP but also to ozone produced during the transport.

32

1 **6 Discussion**

2 The succession of low- and high-pressure systems plays a key role in explaining the day-to-
3 day variations of lower tropospheric ozone over North East Asia. In May 2008, 5 events
4 covering 2-3 days each and leading to significant ozone enhancement in the lower
5 troposphere have been identified. In order to evaluate the regions of influence of the frontal
6 and cyclonic activity on the ozone distribution, we calculated monthly means of lower and
7 upper tropospheric ozone columns (Fig. 9). The monthly means are given with a $0.25^\circ \times 0.25^\circ$
8 horizontal resolution. The upper tropospheric ozone columns are the most affected by the
9 ozone subsiding transfer induced by the tropopause perturbations associated with frontal
10 activity. Looking at UT ozone columns larger than 40 DU provides a view of the region of
11 influence of the frontal and cyclonic activity in terms of ozone enhancement. This region is
12 located north of 40°N and extends from Inner Mongolia to North China and the North of
13 Japan. South of 40°N , the influence of the frontal and cyclonic activity on lower tropospheric
14 ozone decreases.

15 In order to investigate the role of pollution in enhanced lower tropospheric ozone columns
16 observed with IASI, we compare monthly distribution of lower tropospheric ozone columns
17 with the distribution of total CO columns and tropospheric NO_2 columns, often used as
18 anthropogenic sources tracers (Fig.9). The NO_2 tropospheric columns are those observed by
19 the GOME-2 instrument operating on the same satellite platform than the IASI instrument
20 (Boersma et al., 2004) (<http://www.temis.nl/airpollution/no2.html>). All the regions of
21 continental East Asia (NCP, Sichuan Basin, North China...) showing large NO_2 tropospheric
22 columns and then indicating large anthropogenic sources present large total CO columns and
23 also large lower tropospheric ozone columns (Fig. 9). A correlation of 0.62 over the entire
24 domain between IASI lower tropospheric ozone and IASI total CO suggests that
25 anthropogenic sources significantly contribute to the ozone observed in the lower troposphere
26 with IASI. The North China Plain, Yangtze River Delta (near Shanghai) and Hubei province
27 (Wuhan region) are the regions most impacted by pollution according to the satellite
28 observations. Large lower tropospheric ozone columns are observed over North China
29 corresponding to the industrialised Shenyang-Harbin axis also visible in CO and NO_2
30 observations (Fig. 9). However, the ozone plume extends more to the west compared to the
31 CO and NO_2 plumes. This may be explained by the influence of the UTLS, which is larger all
32 over the northern part of the domain. Lower tropospheric columns of ozone might also be

1 overestimated during the retrieval because the region is partly arid. Indeed, the ozone retrieval
2 can be partly impacted in regions of low emissivity. In the southern part of the domain,
3 enhanced lower tropospheric ozone columns are observed in the Sichuan Basin and
4 Guangdong province in coincidence with enhanced CO and NO₂ columns. In this latter
5 region, closer to the equator, the distance between two successive swaths of IASI increases.
6 Then, the spatial and temporal coverage of IASI decreases and it is then less easy to follow
7 the daily variations of ozone. Moreover, the maximum of sensitivity of IASI ozone retrievals
8 in the tropics is usually higher in altitude around 5 km (Dufour et al., 2012). IASI
9 observations are then less suitable to efficiently monitor pollution in such cases.

10

11 **7 Conclusion**

12 Based on ozone and CO retrieval from IASI, we elaborate an analysis method to diagnose
13 which processes contribute to ozone enhancement in the lower troposphere. We apply the
14 method to evaluate the respective role of the stratospheric and the photochemical sources of
15 ozone on the day-to-day variation of the lower tropospheric ozone distribution over East Asia.
16 The study allows us to stress how satellite observations can help in monitoring and identifying
17 these different sources. We focus on late springtime because the cyclonic activity – well
18 known to drive the stratosphere-troposphere exchanges – is important and the photochemical
19 production of ozone in polluted areas can be significant at this time of the year.

20 We demonstrate that ozone profiles and semi-independent ozone columns between the surface
21 and 12 km associated with simultaneous CO measurements from IASI provide a powerful
22 observational dataset to identify the stratospheric and anthropogenic origin of the lower
23 tropospheric ozone. We show that UT ozone columns larger than 40 DU are a proxy to
24 identify the region of subsiding ozone associated with the tropopause perturbation induced by
25 low-pressure weather systems. Combined with LT ozone columns larger of ~30 DU, it
26 identifies the areas in the lower troposphere affected by the UTLS reservoir of ozone. One of
27 the advantages of IASI is to provide 3-dimensional observations of ozone distribution at
28 synoptic scale when cloud free. The analysis of vertical section in longitude or latitude allows
29 one to identify more precisely the areas where the lower troposphere is connected to the
30 UTLS reservoir and the region of possible irreversible stratosphere-troposphere exchanges.
31 On the contrary, we show that large LT ozone columns when not associated with large UT
32 ozone columns but with enhanced CO total columns – used as a pollution tracer – indicate the

1 areas where the photochemical production of ozone forms part of the observed ozone
2 enhancement in the lower troposphere. Once again, the 3D observational capability of IASI
3 (vertical sections) allows one to evaluate if the ozone enhancement observed in the LT is
4 disconnected from the UTLS reservoir and thus to assess the anthropogenic origin of the LT
5 ozone enhancement or the mixing of the sources. We also show that enhancement ratio of O₃
6 to CO, consistent with those from literature, can be derived from IASI.

7 As expected, the succession of low- and high-pressure systems strongly influences the day-to-
8 day variations in lower tropospheric ozone over Northeast Asia during springtime, both
9 leading to LT ozone enhancements. We show that the ozone subsiding transfer due to the
10 tropopause perturbations associated with the low-pressure systems affect the free and lower
11 tropospheric ozone over large regions. We determine the region of influence of such systems,
12 located mainly above 40°N but with some particular intense events (e.g. cut-off low from 11
13 to 13 May 2008) impacting southern regions such as the NCP for few days. The vertical
14 dimension provided by IASI allows the identification of the STE areas, which are located in
15 the southern part behind the cold front in the case of the frontal system and on the southern or
16 south-eastern flanks of the low in the case of a cut-off low. Note that the STE are expected to
17 occur preferentially on the western and southern flanks of the trough.

18 Based on the case of a cut-off low travelling over the NCP from 11 to 14 May 2008, we show
19 that such systems, with potential convective capacity, when they travel over highly polluted
20 regions, play a key role in the transboundary transport of pollutants. We identify from the
21 O₃/CO enhancement ratio estimated from IASI observations that significant ozone
22 photochemical production occurs during the transport from the NCP on 12 May to the Sea of
23 Japan on 14 May.

24 In addition to the stratospheric influence on tropospheric ozone in the northern part of the
25 domain, most of the enhanced lower tropospheric ozone columns are observed in regions
26 mainly impacted by strong pollution level. Significant correlations between CO (used as a
27 pollution tracer) and ozone in the lower troposphere have been found. Moreover, the analysis
28 of vertical sections of ozone concentrations over NCP indicates that ozone concentrations are
29 enhanced only in the lower troposphere in such regions, indicating the anthropogenic origin of
30 the observed ozone enhancements. The maximal values of ozone are observed between 2 and
31 4 km in cases where an anticyclonic situation is well settled over the NCP (e.g. 5 and 15 May
32 2008). This is in agreement with in situ measurements (Huang et al., 2014), considering the

1 limited vertical resolution of IASI and its limited sensitivity to surface ozone. Because of
2 these limitations, it is not possible to determine more precisely the altitude of the ozone
3 enhancements in the troposphere. This is all the more penalizing when stratospheric and
4 photochemical events occur at the same time. The lack of vertical resolution does not allow
5 the various contributions to be differentiated. Combined with modelling studies, advanced
6 satellite products coupling UV and IR information such as the recent IASI+GOME-2 product
7 (Cuesta et al., 2013) as well as the next generation of satellite instruments (Crevoisier et al.,
8 2014, Veeffkind et al., 2012) should help assessing this issue.

9

10 **Acknowledgements**

11 We acknowledge the Institut für Meteorologie und Klimaforschung (IMK), Karlsruhe,
12 Germany, for a licence to use the KOPRA radiative transfer model. This study was supported
13 by the French Space Agency - CNES (project “IASI-TOSCA”). The IASI mission is a joint
14 mission of Eumetsat and the Centre National d’Etudes Spatiales (CNES, France). The IASI
15 L1 data are distributed in near real time by Eumetsat through the Eumetcast system
16 distribution. We acknowledge the Ether CNES/CNRS-INSU database (<http://www.pole-ether.fr>)
17 for providing access to IASI Level 1 data. We acknowledge the LATMOS/ULB for
18 the provision of IASI CO total columns through the Ehter CNES/CNRS-INSU database. The
19 authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision
20 of the HYSPLIT transport and dispersion model and/or READY website
21 (<http://www.ready.noaa.gov>) used in this publication. We acknowledge the free use of
22 tropospheric NO₂ column data from the GOME-2 sensor from www.temis.nl. The
23 ozonesonde data used in this study were mainly provided by the World Ozone and Ultraviolet
24 Data Centre (WOUDC), the Southern Hemisphere Additional Ozonesondes (SHADOZ), and
25 the Global Monitoring Division (GMD) of NOAA’s Earth System Research Laboratory and
26 are publicly available (see <http://www.woudc.org>, <http://croc.gsfc.nasa.gov/shadoz>,
27 <http://www.esrl.noaa.gov/gmd>). The authors thank all those responsible for the WOUDC,
28 SHADOZ, and GMD measurements and archives for making the ozonesonde data available.

29

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1 Table 1. Ozonesonde stations used for the validation. “N days” represents the number of
 2 measurements matching the coincidence criteria.

Station	Location		N days	Station	Location		N days
Ankara	39.97°N	32.86°E	50	Tateno	36.10°N	140.10°E	4
Aquila	42.38°N	13.31°E	11	Uccle	50.80°N	4.35°E	390
Barajas	40.47°N	3.58°W	139	Ushuaia	54.85°S	68.31°W	2
Beijing	39.54°N	117.12°E	7	Valentia	51.93°N	10.25°W	33
Bratts Lake	50.20°N	104.70°W	56	Wallops Island	37.90°N	75.70°W	15
Broadmeadows	37.69°S	144.94°E	19				
Churchill	58.74°N	94.07°W	46	Hanoi	21.02°N	105.80°E	16
De Bilt	52.10°N	5.18°E	104	Hilo	19.43°N	155.04°W	62
Edmonton	53.55°N	114.11°W	2	Hong Kong	22.31°N	114.17°E	93
Egbert	44.23°N	79.78°W	57	Irene	25.90°S	28.22°E	4
Goose Bay	53.31°N	60.36°W	98	Java	7.50°S	112.60°E	6
Hohenpeissenberg	47.80°N	11.00°W	319	Kuala Lumpur	2.73°N	101.70°E	5
Huntsville	34.72°N	86.64°W	9	Nairobi	1.27°S	36.80°E	78
Kelowna	49.93°N	119.40°W	124	Naha	26.20°N	127.70°E	0
Lauder	45.04°S	169.68°E	5	Natal	5.49°S	35.80°W	64
Legionowo	52.40°N	20.97°E	133	Pago	14.23°S	170.56°W	13
Lindenberg	52.21°N	14.12°E	148	Panama	7.75°N	80.25°W	2
Macquarie Island	54.50°S	158.94°E	1	Reunion	21.06°S	55.48°E	87
Payerne	46.49°N	6.57°E	389	Samoa	14.23°S	170.56°W	3
Praha	50.01°N	14.45°E	143	San Cristobal	0.92°S	89.60°W	24
Sapporo	43.10°N	141.30°E	12	Santa Cruz	28.46°N	16.26°W	2
Stony Plain	53.55°N	114.11°W	57	Watakosek	7.50°S	112.60°E	16

3

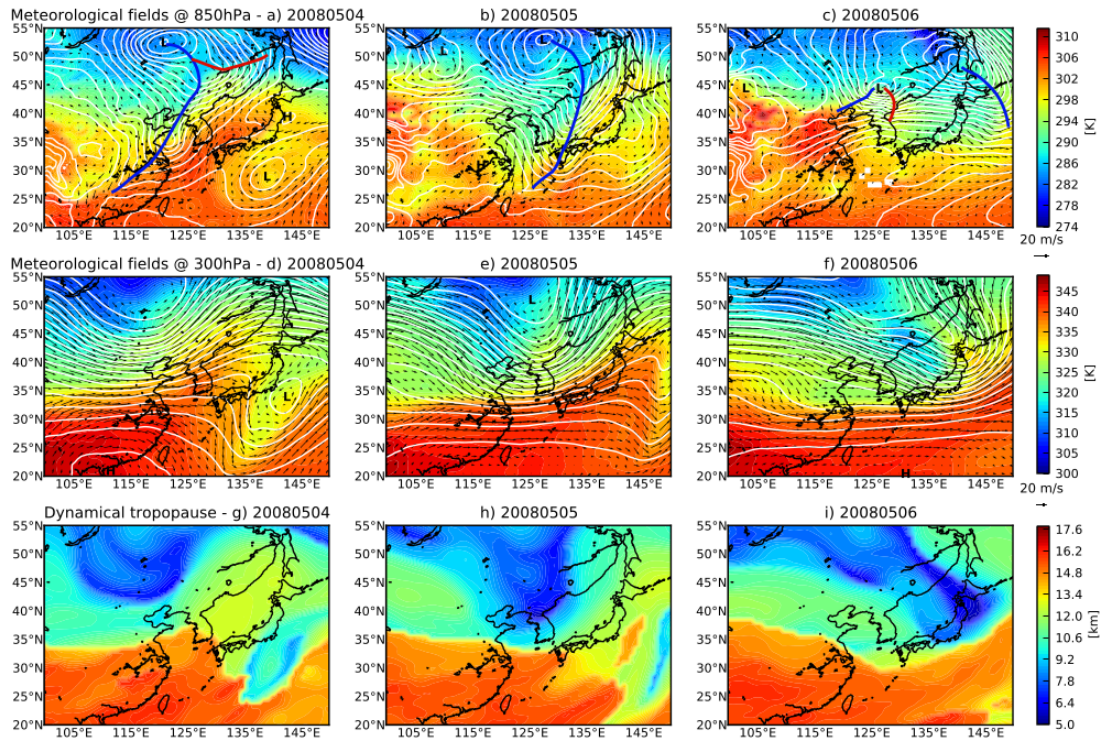
Station	Bias	RMS	R
All stations	-0.6 (2.8)	2.8 (13.7)	0.70
East Asia	-2.2 (-9.5)	2.7 (11.6)	0.70
Beijing	-2.6 (-9.0)	2.6 (9.0)	0.71
Sapporo	0.8 (3.9)	3.9 (19.8)	0.68
Tateno	-2.6 (-12.1)	2.3 (10.8)	0.60
Hong Kong	-2.6 (-10.9)	2.2 (9.6)	0.67

4

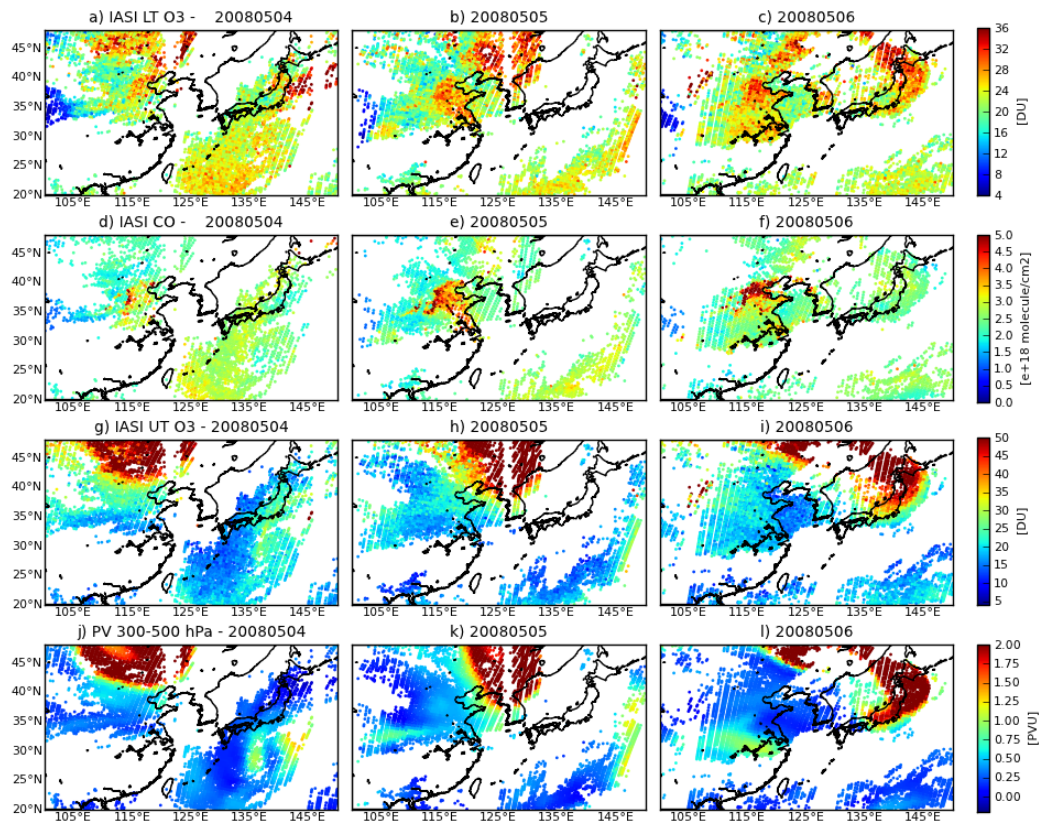
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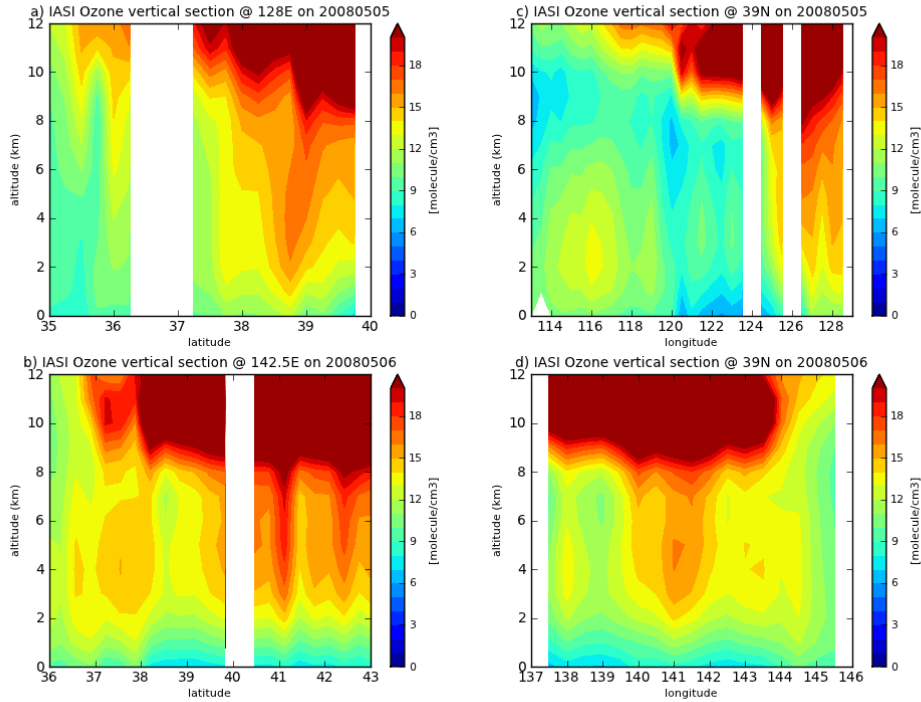


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 2 Figure 1. Meteorological situation given at (a-c) 850 hPa and (d-f) 300 hPa from 4 to 6 May
 3 2008 as well as (g-i) the dynamical tropopause. All the meteorological variables are derived
 4 from the ERA-Interim reanalysis. The color filled contours in (a-f) represent the equivalent
 5 potential temperature and the white contour the geopotential height. The “L” and “H”
 6 symbols represent the centre of lows and highs respectively. Horizontal winds are also
 7 plotted. The cold and warm fronts are displayed in blue and red respectively on the top panel.



1

2 Figure 2. (a-c) Lower tropospheric ozone columns (surface to 6 km asl) retrieved from IASI
 3 from 4 to 6 May 2008; (d-f) Total CO columns retrieved from IASI; (g-i) Upper tropospheric
 4 ozone columns (6 to 12 km asl) retrieved from IASI; (j-l) Potential Vorticity (PV) from ERA-
 5 Interim reanalysis averaged between 300 and 500 hPa.



1

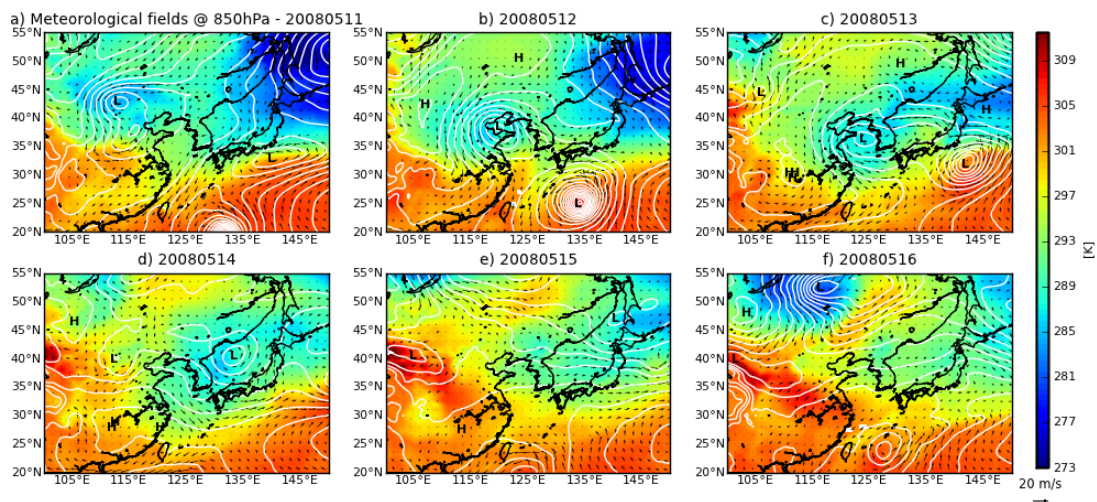
2 Figure 3. Vertical section of ozone concentration (in molecule/cm³) retrieved from IASI along
 3 specific longitudes – (a) 128°E on 5 May 2008, (b) 142.5°E on 6 May 2008 – and along
 4 specific latitudes – 39°N on 5 May 2008 (c) and 6 May 2008 (d). The longitudinal
 5 (latitudinal) sections are computed over 1° around the specific longitude (latitude) with a
 6 0.25° resolution in latitude (longitude).

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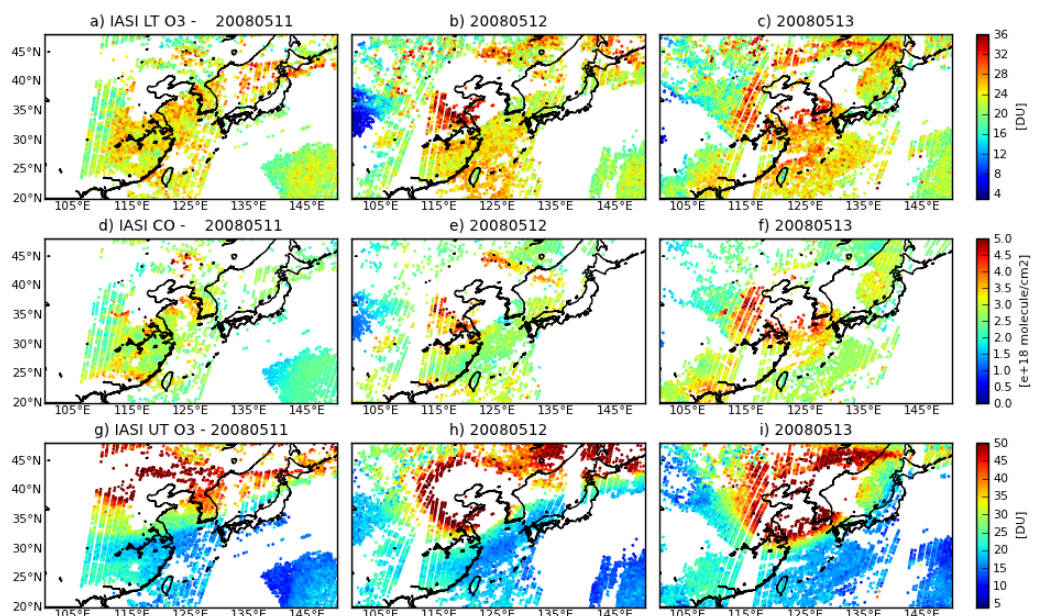
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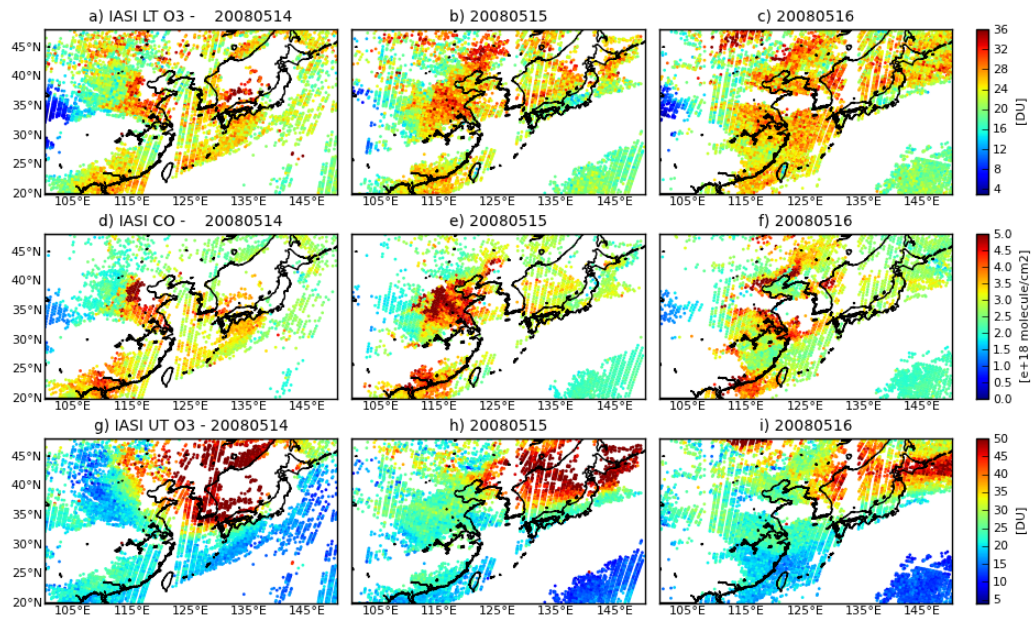
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Figure 4. Meteorological situation given at 850 hPa from 11 to 16 May 2008. The color filled contours represent the equivalent potential temperature and the white contour the geopotential height. The “L” and “H” symbols represent the centre of lows and highs respectively. Horizontal winds are also plotted.

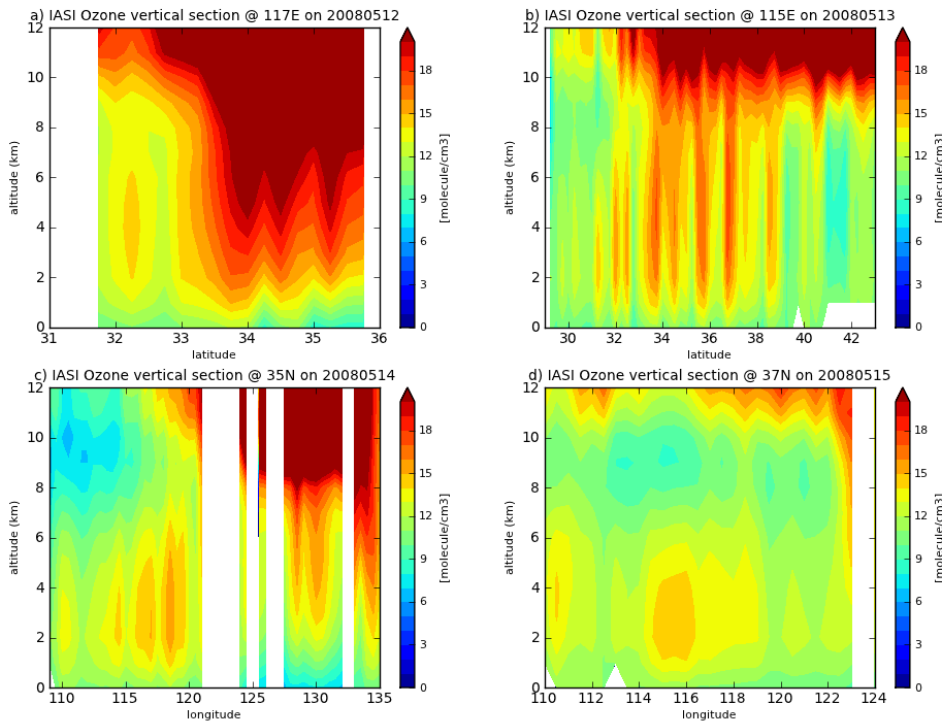


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Figure 5. (a-c) Lower tropospheric ozone columns (surface to 6 km a.s.l) retrieved from IASI from 11 to 13 May 2008. (d-f) Total CO columns retrieved from IASI. (g-i) Upper tropospheric ozone columns (6 to 12 km asl) retrieved from IASI

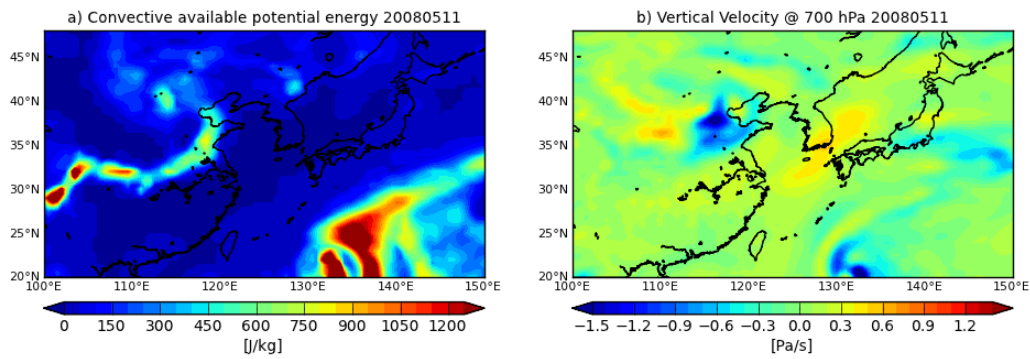


1
2 Figure 6. Same as Fig. 6 for 14 to 16 May 2008.

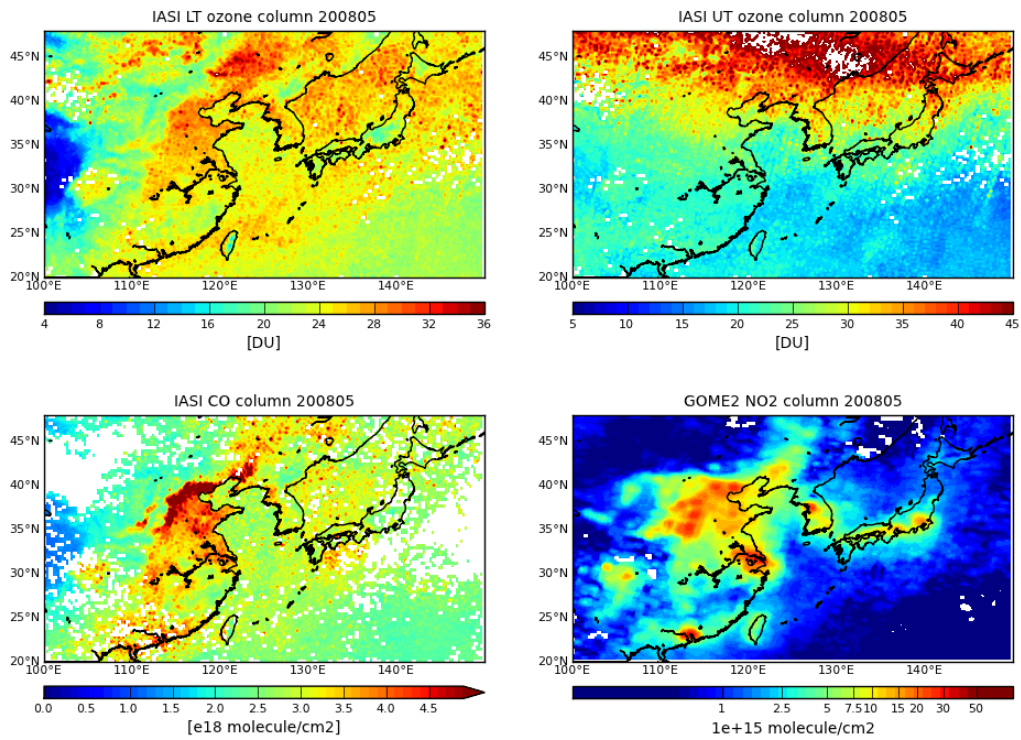


3
4 Figure 7. Vertical section of ozone concentration (in molecule/cm³) retrieved from IASI along
5 specific longitudes – (a) 117°E on 12 May 2008, (b) 115°E on 13 May 2008 – and along
6 specific latitudes – (c) 35°N on 14 May 2008, (d) 37°N on 15 May 2008. The longitudinal

1 (latitudinal) sections are computed over 1° around the specific longitude (latitude) with a
 2 0.25° resolution in latitude (longitude).

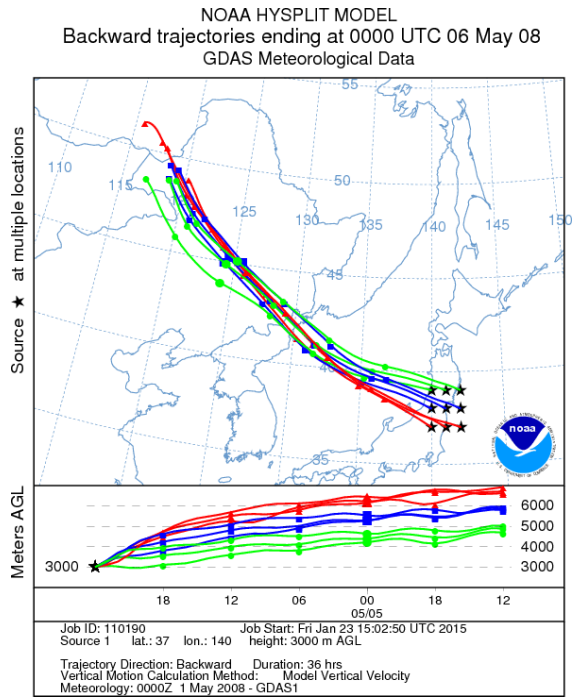


3
 4 Figure 8. Convective available potential energy (a) and vertical velocity at 700 hPa (b) from
 5 ERA-Interim reanalysis.



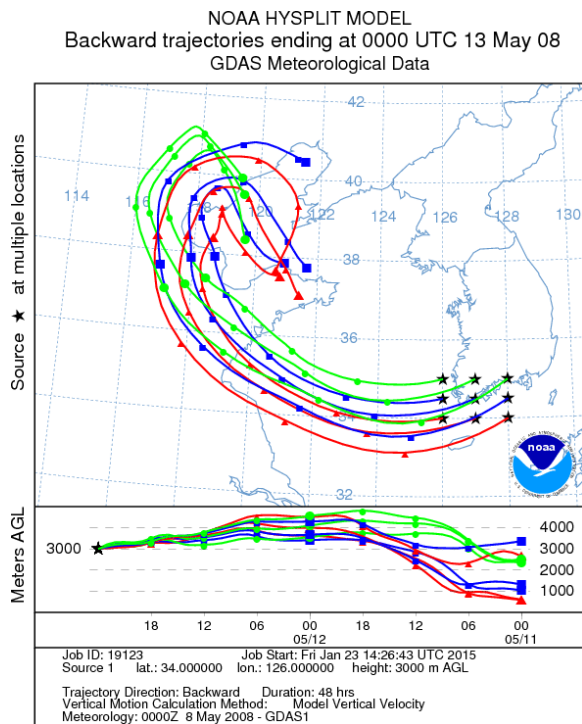
6
 7 Figure 9. Monthly lower (upper left) and upper (upper right) tropospheric ozone columns
 8 observed by IASI in May 2008 as well as monthly IASI total CO columns (lower left) and
 9 GOME-2 NO_2 tropospheric columns (lower right) observed in May 2008. The average is
 10 calculated for a $0.25^\circ \times 0.25^\circ$ resolution grid.

1 Supplementary material



2

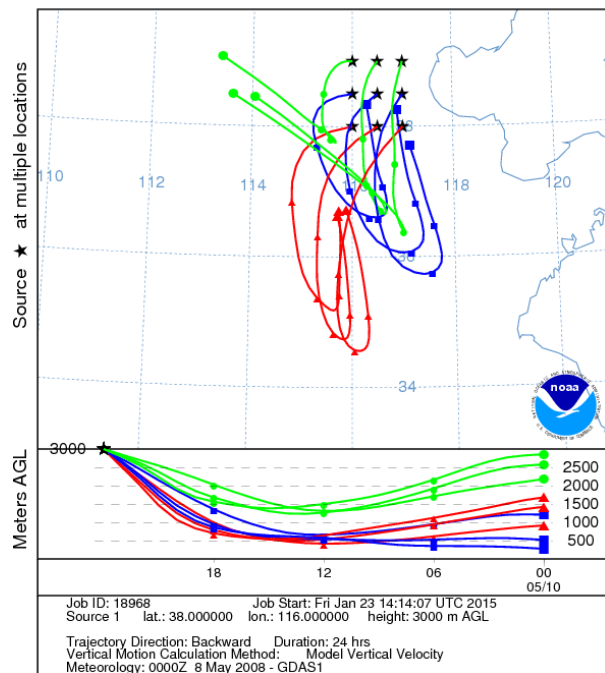
3 Figure S1. 36-hours backward trajectories ending at 3km on 6 May 2008 in the region of
4 Tokyo.



5

6 Figure S2. 48-hours backward trajectories ending at 3km on 13 May 2008 in the region of
7 South Korea.

NOAA HYSPLIT MODEL
 Backward trajectories ending at 0000 UTC 11 May 08
 GDAS Meteorological Data



- 1
- 2 Figure S3. 24-hours backward trajectories ending at 3km on 11 May 2008 in the region of
- 3 Beijing.
- 4