Ground-Based Remote Sensing of the ABL Structure in Moscow and Its Use to Estimate Pollutant Surface Emission Rates

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(Manuscript received 21 December 2012, in final form 16 December 2013)

ABSTRACT

Currently used methods to estimate surface pollutant emissions require a set of specific air-sampling surveys. Data from a network of ground-based sodars and a network of air-quality stations in Moscow, Russia, are used to estimate the emission rates of carbon monoxide (CO) and nitric oxide (NO). The sodar network, consisting of three “LATAN-3” Doppler sodars and three “MTP-5” microwave temperature profilers, is used to measure the vertical profiles of vertical and horizontal wind velocity, wind direction, and temperature, which are used to determine the average mixing-layer height. The network of ground-based air-quality stations, consisting of 17 automated stations distributed uniformly across Moscow, continuously measured the CO and NO concentrations. This study focuses on an anticyclonic episode of high surface pressure over Moscow during 30 July–1 August 2012. After sunrise, the solar-induced convection effectively moderated the pollutant levels in the lowest 100–200 m. After sunset, convective mixing stopped and the wind weakened, which allowed CO and NO to reach hazardous levels. With an assumption of an average mixing-layer height of 150 m, the resulting estimate of surface emission of CO is \( \frac{6}{m^2 s} \), whereas that for NO is \( \frac{0.6}{m^2 s} \).

1. Introduction

Short-range transport of pollutants depends on the wind field and the intensity of turbulent mixing in the atmospheric boundary layer (ABL; Monin and Yaglom 2007). As a result, to understand local pollutant transfer and dispersion over a city, researchers should know the wind field, the characteristics of the pollutant sources (including their positions, temperatures, and emission rates), the turbulent characteristics, and the pollutants’ deposition rates. In addition, to assess the consequences of urban air quality, one has to estimate the pollutant concentrations in the urban layer from the surface up to 50–100 m. Stratification of this layer strongly influences the pollution transport, with stable stratification worsening the air quality by suppressing turbulence and convection and unstable stratification having the opposite effect.
Moscow, Russia, usually has sufficient wind to keep the pollutant concentrations at safe levels. In anticyclonic and similar meteorological situations with weak wind, however, the heavy vehicle traffic in Moscow causes pollutants to accumulate near the surface. Within a few hours of such conditions, the carbon monoxide (CO) and nitric oxide (NO) concentrations can exceed their threshold limit values (TLV) for short-term exposure. The TLV for CO is 4000 μg m\(^{-3}\), and that for NO is 200 μg m\(^{-3}\).

Sonic detection and ranging (sodar) data provide information about wind speed, wind direction, and the intensity of turbulent mixing in the ABL (Kallistratova 1994; Emeis et al. 2007). Sodar-derived vertical velocities also provide a measure of convection intensity. For urban measurements, sodar has advantages over radar and lidar wind profilers. Unlike radar, it is not necessary to allocate an electromagnetic band for sodar, and relative to low-power lidar a sodar has a greater height range (higher-power lidar has greater range but requires further governmental approval). Moreover, the cost of a sodar unit is typically about one-half that of radar and lidar.

For the coherent structures (cells) formed by convection, sodar of the kind used here can provide details of the air up to a height of about 1 km (e.g., Granberg et al. 2009). The vertical dimension of each structure is roughly equal to the height of the convective layer, and they have an aspect ratio (vertical:horizontal) of about 1:2–1:4 (e.g., Mikhailova and Ordanovich 1991; Lappen and Randall 2005; Granberg et al. 2009). In terms of their inner structure, large convective structures in the shape of a toroid have been observed by a sodar network in the Kalmykian desert (Granberg et al. 2009; Kalmykia is part of the Russian Federation), which is an area of higher heat flux than Moscow; nevertheless, we expect the convective structures in Moscow to be toroidal.

In the ideal case, a database with the characteristics of pollutant sources should be made to provide necessary input for models of short-range transfer of pollutants in a city air basin. Each source’s classification, inventory, and parameterization should be stored in a database, similar to that constructed by Zakarin and Kramar (1991) and as recommended in Bluett et al. (2004). Making, using, and updating such a database is a very expensive and time-consuming process, however. In Moscow, it may be useful to use an existing air-monitoring network for the pollutant concentrations at the surface and then to derive current characteristics of the sources from consideration of the wind structure of the overlying urban layer.

Here we use sodar data together with ground-based air-quality sensor data for a high pressure episode over 30 July–1 August 2012 to determine the wind structure in the Moscow-area ABL and to estimate the amount of CO and NO emissions. For this estimate, we use a simple box model. Although more detailed models of urban air pollution exist, a box model is considered to be adequate to address the present goal, which is to quantify average emission rates of specific air pollutants in Moscow. We estimate an average CO emission rate of \(\sim 6 \mu g m^{-2}s^{-1}\) and an average NO emission rate of \(\sim 0.6 \mu g m^{-2}s^{-1}\). These estimates should be viewed as first attempts to understand how pollution from vehicles affects the atmospheric environment in Moscow.

2. Ground-based remote sensing and the air-quality network in the Moscow metropolitan area

To acquire data on parameters that affect short-range transport and dispersion of pollutants in the Moscow region, the Obukhov Institute of Atmospheric Physics (IAPh) of the Russian Academy of Sciences, together with the Physical Faculty of Moscow State University (MSU), designed an experimental network of ground-based remote sensors to monitor ABL parameters. The network included sodars and passive microwave profilers.

“LATAN-3” sodars (Kouznetsov 2009), designed and produced at IAPh, were placed at three sites: 1) in the center of Moscow (IAPh), 2) on the periphery of the city (MSU), and 3) in a rural area 45 km west of Moscow at the Zvenigorod Scientific Station (Zvenigorod) of IAPh (Fig. 1). Each unit sequentially emits acoustic probing from three transmitters and processes the received echo signals. After processing three sequential echo signals, the unit determines the instantaneous vertical profiles of the three components of wind velocity. The profile ranges from the surface to a height of \(~200–800\) m, depending on the surrounding acoustic noise, with a vertical increment of 20 m. Instantaneous values are averaged over 30 min, and each unit provides average vertical profiles that are freely available in real time on the IAPh (http://devio.us/~roux/IFA/) and Zvenigorod (http://devio.us/~roux/ZVE/) Internet sites.

The “MTP-5” passive microwave profilers measured vertical profiles of air temperature from the surface to a height of about 600 m with a vertical increment of 50 m and a data-averaging time of 5 min [see Kadygrov et al. (2003) for further details about MTP-5]. The profilers were at Zvenigorod and MSU and also at the Hydro-meteorological Centre of Russia (Federal Service for Hydrometeorology and Environmental Monitoring), which is 3.7 km northwest of the IAPh.

The surface concentrations of various pollutants, including CO and NO, were measured by the Mosecomonitoring network of automatic stations, arranged by the city council of Moscow (data were obtained from the State Environmental Institution: Mosecomonitoring...
The stations are equipped with certified automatic gas analyzers and other instruments that run continuously [see Gorchakov et al. (2006, 2011) for detailed descriptions of the equipment]. The station locations are shown in Fig. 1 and are described in appendix A.

3. The high-pollution episode of 30 July–1 August 2012 in Moscow

As an indicator of surface pollution, we selected CO because of its low chemical reactivity and its being one of the main emissions from vehicles. For comparison, we also analyze the highly reactive NO species, which primarily comes from vehicle exhaust. Because of the low vehicle speeds and frequent traffic jams, vehicles are a major source of air pollution in Moscow.

a. General conditions

High CO concentrations occur mainly under conditions of a low pressure gradient or an anticyclonic weather system. These conditions have weak winds and clear skies, allowing the solar radiation to stimulate convection. Because of the lack of precipitation that leads to wet deposition, the pollutants are not washed out. Because our aim was to use the sodar method
during a period with dangerously high CO levels, we chose to analyze an anticyclonic weather episode that lasted for several days. Such an episode occurred on 30 July–1 August 2012.

In such an anticyclonic weather episode, the solar heating of the surface during daytime generates convective mixing, which reduces the CO levels at the surface as cleaner upper-level air is brought down. After sunset, this air-cleaning mechanism shuts off as the wind weakens, meaning that the pollutants from near-surface sources can accumulate. As a result, the surface pollutant concentrations can quickly increase, often reaching hazardous levels.

Because we are focused on mesoscale dispersion, we will assume that the CO concentration within the city depends only on emission, transfer, and deposition processes. Because CO is relatively unreactive, this is a good assumption for CO. As a simple comparison, we also use the same model for the highly reactive and highly hazardous NO. To put the concentrations into clearer comparison, we normalize each concentration to the TLVs for CO and NO are 4000 and 200 μg m\(^{-2}\), respectively. The typically observed concentrations of these pollutants at the urban stations range within 0.02–0.1 of the TLV.

In this study, we neglect the influence of long-range pollutant transfer on the city air pollution level. This neglect is justified by observations at the background monitoring station in Zvenigorod, where the background pollutant levels are lower by at least a factor of 10 than those simultaneously measured at city stations. The resulting 10% correction is insignificant when compared with other assumptions and approximations in the model.

**b. Time dependence of CO and NO**

The normalized CO and NO concentrations have a similar diurnal trend. Figures 2 and 3 show their concentrations during the episode periodically rising up to near the TLV after sunset and then falling back down after sunrise. During the episode, which started at around 2000 LT (UTC + 4 h) on 30 July and lasted until the daytime on 1 August 2012, the lowest values of concentrations were in the typical range (i.e., 0.02–0.1 of the TLV).

The consistent rise–decay behavior of CO and NO supports the solar-heating–convective-mixing mechanism described above. The simultaneous growth and following fall of CO and NO concentrations at all monitoring stations indicates a widespread rather than local cause. Moreover, the peaks after sunset occur despite the relative decrease in vehicular traffic. Thus, the difference in chemical reactivity between CO and NO does not lead to differences in their global (within the city) diurnal trends.

The relatively low daytime values are a direct consequence of the mixing effect of intensive convection. Then at night, the concentrations reach their maxima because of the weak wind and an absence of convective mixing. As the episode progressed, the maximum values approach, and then exceed, the TLV as the average horizontal wind speed decreased. These speeds decreased from 2 m s\(^{-1}\) on the first night to 1 m s\(^{-1}\) on the second night and to 0.5 m s\(^{-1}\) on the last night (see Fig. 3). This very small horizontal wind provides the sole mechanism for nocturnal surface air clearing during such episodes.

Even during the day, however, the convection mechanism has a limited effectiveness at clearing surface air. For example, Fig. 2 shows that the daytime concentration values hold approximately steady at nonzero values. Particularly high values are at station 14, where the CO level remained above 0.4 of the TLV. The high daytime levels occur because the pollution source is strong nearby: the station lies near an intersection of five high-traffic highways.

The surface pressure map at 0000 UTC 31 July 2012 shows Moscow in a pole of high pressure. For the overnight of 30 July, meteorological station 27612 [~1 mi (1.6 km) northeast of site 01 in Fig. 1], reported clear sky, haze, and a horizontal wind of 0–1 m s\(^{-1}\). Figure 3 shows the supporting sodar and pressure data from the same station.

Despite the weak mean horizontal wind, the urban surface layer during daytime on 30 and 31 July was efficiently ventilated by convection (Figs. 2 and 3). According to the sodar data in Fig. 4, the convection produced updrafts slightly exceeding 1 m s\(^{-1}\) and similar-magnitude downdrafts. The daytime updrafts often extend up to 300m and last for over 300 s; thus, convective mixing during the day could effectively mix the CO and NO (and any other pollutant) up to at least 300 m.

On 31 July, the increase in CO and NO concentrations began after sunset (2140 LT), which is still within the rush hour (~1600–2200 LT). Convection ceases at this time, and stable stratification sets in, with horizontal wind speeds in the ABL dropping below 1 m s\(^{-1}\) (see Fig. 3 and temperature and wind profiles at 2200 LT in Fig. 5). This decrease in horizontal wind speed correlates with a sharp increase in the CO and NO concentrations (Figs. 2 and 3). The sodar-derived wind speeds in Figs. 3–5 are from central Moscow, but the same decay in wind speed was also found from the rural sodar site, marked “Zvenigorod” in Fig. 1, outside of Moscow.
Thus, the conditions were favorable for the accumulation of pollutants over a large horizontal scale.

c. Surface emission intensity

The measured near-surface concentrations, together with the information from the sodar network and temperature profilers, are used here to estimate the upper-bound surface source emission rates. We assume that, at the maximum of the surface concentration of the pollutant, emission – removal is in dynamic equilibrium. We then apply a simple, balanced box model (e.g., Zakarin and Kramar (1991) or Bluett et al. (2004) or appendix B) to the Moscow area. Thus, over the time in which it takes the horizontal wind to cross Moscow (approximately 50 km across), an entire volume of pollutants over Moscow will be carried away through the city boundaries and will be replaced by freshly emitted pollutants. For our anticyclonic episode, we use

![Figure 2: CO concentrations from 30 Jul to 1 Aug 2012 for two sets of stations, as numbered in Fig. 1: (a) a set of sites placed at M/DS and SHW only and (b) RD and natural area stations only (should have lower level of pollutants elsewhere in the city). Time shown is LT (UTC + 4 h). Values are normalized to the TLV for short-term exposure, which is 4000 μg m⁻³ for CO. The arrows designate sunrise (0530) and sunset (2140) times.](image-url)
the sodar-derived horizontal wind speed of 1 m s\(^{-1}\) and
the average mixing-layer height of 150 m (Fig. 5). The
latter height is identified as the boundary between
polluted air and the clean air brought from the sur-
roundings by a strong upper-level wind. The level should
be slightly higher than the height at which the horizontal
wind speed begins to rapidly increase with height. We
confirmed this mixing-layer height by comparing the
observed temperature gradient with the dry-adiabatic
temperature gradient. Thus, our conceptual box of pol-
luted air has a volume of about \(3 \times 10^{11} \text{ m}^3\) and a time for
air to cross the Moscow area of \(5 \times 10^4 \text{ s} = 14 \text{ h}\).

To simplify the estimate, we assume that the pollut-
ants in the mixing layer are distributed uniformly. Measurements support this assumption: Gorchakov et al. (2011) measured concentrations of CO at site 01
at heights 2, 130, 250, and 350 m and found average
values of 560, 390, 400, and 370 \(\mu\text{g m}^{-3}\), respectively.
For the episode of 31 July 2012, the average CO con-
centration in the layer is approximately 0.5 of the TLV
or 2000 \(\mu\text{g m}^{-3}\). Then, the overall quantity of CO
emitted into the Moscow area during the 14-h interval
is \(~600 \text{ t}\). If one approximates Moscow as a circle 50 km
across, the resulting estimate of emission rate of CO
from the distribution of sources is \(q_{\text{CO}} \sim 6 \mu\text{g m}^{-2} \text{s}^{-1}\).
A similar estimate for NO gives the much smaller
\(q_{\text{NO}} \sim 0.6 \mu\text{g m}^{-2} \text{s}^{-1}\).

The above estimates assume a constant concentration
to the top of the mixed layer. If there was instead a linear
decrease of concentration to zero at the layer top, the
resulting estimates of pollutant emission rate would be
one-half as large. The above emission-rate estimates are
thus likely upper bounds.

An alternative method would be to divide the regions
into smaller boxes (grid cells) having different concen-
tration and then to separately estimate their emission
rates. For example, inspection of Fig. 2a suggests that
the area around station 14 experienced an emission rate
of about 2 times the Moscow average value at night and
considerably more than 2 times in the daytime.
Assume at first that there is no vertical removal of pollutants. Following the previous analysis, a similar calculation gives a concentration of CO of 0.4 of the TLV or 1600 μg m\(^{-2}\) s\(^{-1}\).

d. Surface emission near the traffic center

We now estimate the daytime surface emission for the traffic center, the crossroads of five highways and other minor roads near site 14. For 31 July, Fig. 2 shows the daytime CO concentration over this region to be constant. The constant condition indicates that emissions are in dynamic equilibrium. As before, we assume for this region a 150-m well-mixed-layer height (slightly higher than the distance from the surface up to the level of a sharp increase in horizontal wind velocity \(V\) in Fig. 4), a 2 m s\(^{-1}\) wind velocity into the layer, and an average concentration of CO of 0.4 of the TLV or 1600 μg m\(^{-3}\) (Fig. 2). Outside the volume of polluted air, the CO concentration is lower by a factor of 10 (see Fig. 2) and thus may be neglected. To simplify the estimate, we assume a horizontal dimension equal to that of a convective cell (or any integer number of cells) that completely mixes the layer over the traffic center. The vertical size of the cell, to the altitude at which the vertical velocity becomes weaker, is about 300 m in Fig. 4. The aspect ratio (height/width) of the cell is about 1/4, meaning the horizontal dimension is about 1200 m. Assume at first that there is no vertical removal of pollutants. Following the previous analysis, a similar calculation gives \(q_{CO} \sim 400 \mu g m^{-2}s^{-1}\). If we instead assume the volume over this traffic center to be a 2 × 2 cluster of convective cells, with all other conditions the same, then \(q_{CO}\) will be one-half as large.

Analogous CO behavior occurs at site 11, which is near the intersection of two highways with a viaduct. The structure of the intersection is simpler than that of 14. Because the daytime concentrations of CO are lower by a factor of 2.5–3 than those for 14, the resulting CO surface emission rates should be about 130–160 μg m\(^{-2}\) s\(^{-1}\).

e. Vertical removal of pollutants from the traffic center

Lappen and Randall (2005) and Granberg et al. (2009) gave the correlation functions for the convective cell structure. Use of their functions shows that the horizontal extent of the central updraft region of the cell is 25%–30% of that of the cell, or about 8% of the area occupied by the cell. With a horizontal wind speed of about 2 m s\(^{-1}\), the volume of the polluted air having a height of 150 m will be removed horizontally in 600 s. Because the updraft speed during this time is about 1 m s\(^{-1}\), in 600 s a volume of a column of 600-m height of polluted air (i.e., 4 times the height of the polluted air layer) will be fully removed vertically from the polluted layer and replaced with clean air from the upper layer. The volume of the tall column is 32% of the polluted air volume (8% of the area multiplied by the 4-times-as-large height of the polluted air layer), and therefore the vertical removal is 32% of the horizontal removal.

f. Comparison with modeled anthropogenic pollutant emission

Here we use the Emissions of Atmospheric Compounds and Compilation of Ancillary Data (ECCAD) Internet application (http://eccad.sedoo.fr/) to compare our result with estimates of anthropogenic emission of CO from several datasets. We used the “web” interface to select the rectangle, consisting of 20–25 pixels, that includes Moscow. We then selected the pixel with maximum emission rate. The space resolution proposed by the ECCAD interface is 0.5°, and emission intensity is averaged over a month or year (depending on the dataset). Using the Monitoring Atmospheric Composition and Climate (MACC)/Megacity–Zoom for the Environment (CityZEN) European Union projects (MACCity) emissions dataset, the CO anthropogenic emission rate for Moscow for July 2009 was likely about 3.1 μg m\(^{-2}\) s\(^{-1}\). Using the regional Netherlands Organisation for Applied Scientific Research (TNO)-MACC gridded anthropogenic emission database for 2003–07, the emission rate for Moscow in 2007 was 18 μg m\(^{-2}\) s\(^{-1}\). Using the Emission Database for Global Atmospheric Research, version 4.2 (EDGARv4.2), from 1970 to 2008, we get an emission rate of 8.9 μg m\(^{-2}\) s\(^{-1}\) for Moscow in 2008. These values are consistent with our estimate of 6 μg m\(^{-2}\) s\(^{-1}\).

To compare the rate with that from other urban areas, for London in 2009 July the MACCity dataset for
anthropogenic CO gives 0.9 $\mu g m^{-2} s^{-1}$, and for New York it gives 2.7 $\mu g m^{-2} s^{-1}$. The same procedure for EDGARv4.2 gives 42 $\mu g m^{-2} s^{-1}$ for New York in 2008 and 1.8 $\mu g m^{-2} s^{-1}$ for London in 2008. Thus, at least during the high-pollution episode analyzed here, the CO emissions in Moscow are close to those in London and New York.

\textbf{g. Comparison for the traffic center near site 14}

A simple ordinary estimate of the emission rate is as follows. The region near site 14 had five highways, each with five traffic lanes in each moving direction. A 1-km$^2$ area in this region includes about 2000 vehicles, each moving at average speed of 10 km h$^{-1}$ with an average emission of 25 g km$^{-1}$ of CO. Each vehicle occupies about 15 m of a traffic lane. We neglect traffic on minor roads. The resulting estimate gives an emission rate of 120 $\mu g m^{-2} s^{-1}$. The initial data for current estimation and dimension of area are somewhat arbitrary. Therefore, the agreement with our estimate calculated in section 3d (qCO $\sim 400 \mu g m^{-2} s^{-1}$) is realistic.

\textbf{4. Conclusions}

We found that the IAPh sodar in central Moscow can, despite the widespread noise, successfully monitor the
The continuity equation in the integral form for any pollutant is used, with integrals calculated over the volume of a box $B$. By Gauss’s theorem, the volume integral over the divergence term changes to the surface integral around $B$. If $c$ is the mass concentration of a pollutant in volume $db$, then

$$\frac{\partial}{\partial t} \int_B c \, db + \oint_{\partial B} \mathbf{v} \cdot \mathbf{n} \, ds = \int_B q \, db,$$

where $\mathbf{v}$ is the velocity of the volume, $ds$ is the surface element of total surface $S$ over the total volume $B$, and $q$ is the sources minus sinks in $db$. The equation means that the change with time in the amount of a pollutant inside the box must equal the difference between the pollutant sources and sinks. We use the box-modeling approach to find the near-surface pollutant fluxes with measured pollutant concentration in the box [i.e., use simplest form of inverse modeling, such as described in Brasseur and Jacob (2013)].

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