Abstract. Air quality, ozone depletion, and climate change are the most important global issues facing society. RADO – Romanian Atmospheric research 3D Observatory is a new, state-of-the-art infrastructure dedicated to research of processes and compounds in the atmosphere. The novelty of this Observatory is the synergetic use of optoelectronic instruments for the survey of the atmosphere from ground-level to the stratosphere. RADO is based on 3D network instrumentation both for gases and aerosols monitoring in order to assess the important volume processes. This paper presents the architecture of the Romanian Atmospheric Observatory and correlation of modelling and experimental data collected at the 5 lidar-based stations in Romania.

Key words: remote sensing, greenhouse gases, aerosols, lidar, observatory.

1. INTRODUCTION

Due to dramatic climate changes, and especially to more frequent extreme events, during the last decades, worldwide Environmental Research concentrated its efforts toward development of climate change prediction and scenarios for improvement of regional climate change predictions. In order to accomplish this objective, a large amount of experimental data must be collected, processed, evaluated and analyzed. These data must also have a good temporal and spatial coverage to be relevant for regional models. At first sight, a lot of experimental techniques are available today to perform this task. Unfortunately, there are some problems in correlating these data: different measurement/processing methods give different results (so that inter-calibration and systematic validation of equipments is necessary), data collection and handling procedures are dramatically different in
terms of measurement and response time, resolution and accuracy, derived parameters are inter-dependent etc. But most critical, few research facilities in the world are able to perform measurements for all important parameters at the same time and approximately in the same location, to fully characterize the air column and its dynamics. Being so rare provided data are not significant at continental scale and therefore cannot improve modelling tools. Satellite instruments, on the other hand, see large areas, but their temporal resolution, along with the spatial resolution in the lower troposphere, where the most important anthropogenic influence, is not enough to assess regional or fast changes such as enhancing of the mid-latitude cyclones activity, frequent floods, droughts, thunderstorms, incidentally tornadoes or the impact of pollution on air quality.

The high variability of physical and optical properties of particles and gases – linked to meteorological parameters such as temperature, humidity and wind speed – require studies of the components in their natural state. Over the past several years, atmospheric features have been intensively investigated through numerical modeling [1] and thanks to experiments [2] detailed information on atmospheric component’s physical and optical properties can be provided by ground-based in situ measurements. Unfortunately these observations often remain limited to the ground. However, ground level concentrations can also be influenced by vertical transport induced by turbulent mass flux. Even more importantly, the direct and indirect climate impact of aerosols depends on the total aerosol load in the atmospheric column and its vertical distribution. Laser remote sensing (lidar) technology turned out to be an appropriate tool in the determination of particle optical properties [4, 5], and ozone concentrations either from ground, airplanes or space. Not only that vertical distribution of atmospheric components can be measured, but their temporal dynamics can be assessed by long-term monitoring, since the response of the instrument is real time. Main drawbacks of ground-based lidars are local operation (and therefore relevance) coupled with a sparse territorial distribution and the limited information that can be directly derived. Beyond the difficulties in calibrating a lidar system (Mie, Raman or water vapor channels), complementary information is required in order to quantitatively derive optical parameters.

2. METHODOLOGY

RADO – Romanian Atmospheric research 3D Observatory is a new, state-of-the-art infrastructure dedicated to research of processes and compounds in the atmosphere. The novelty of this Observatory is the synergetic use of optoelectronic instruments for the survey of the atmosphere from ground-level to the stratosphere.

RADO has three main components: the Observation Network, Data Center and Science Center. It is proposing 3D network instrumentation both for gases and aerosols monitoring in order to assess the important volume processes. Remote
sensing instruments (lidars) are used together with integrated column sensors (sun tracking photometers) and in situ ground-level monitors (for gases and particles). Lidars are profiling aerosols and water vapor up to the troposphere, so that layer altitudes can be derived. Using 7-9 wavelengths, sun photometers provide more optical parameters, but integrated in the air column. Data obtained from ground-level monitors are used as ground-truth and calibration values. This synergy of point monitor-total column and profiles supplemented by modelling tool and satellite data is the originality of the RADO giving a unique potential for real atmospheric monitoring and assessment solutions long term platform.

The Observation Network is based on 5 existing lidar stations, which now operate as the Romanian Lidar NETwork (ROLINET). Four of the five sites are equipped with elastic backscatter & depolarization lidars, having a dynamic range from 500m to 15km and a spatial resolution of 15m. A multiwavelength Raman lidar (3 elastic + 2 Nitrogen Raman + 1 water vapor channels) and an AERONET sun photometer are used at Magurele super site to derive independent aerosol optical properties.

Lidar systems use a laser to excite particles of the atmosphere. Detected signal is generated by air density fluctuations (Rayleigh scattering) and by small aerosol particles always present in the atmosphere. The presence of aerosol particles causes an increasing of the backscattered light and thus it can be detected knowing the clean atmosphere background. The optical signals received by the telescope are selected by wavelength using an optical analyzer and then directed to the photosensitive detectors. These will convert the optical signal into electrical signal, recorded as a function of time by analog-to-digital converters and/or photon counting devices and the results are stored onto a computer. The main advantage of a lidar system is that it provides real-time profiles of atmospheric components, which makes it useful for troposphere and even stratosphere study.

For each of these stations, the infrastructure is improved by implementing new state-of-the-art sensors (O3 and CO2, monitors, particle sizers, UV cameras and IR cameras for 3D imagery, sunphotometers), part of these being developed at NILU (Norwegian Air Research Institute).

A commercial off the shelf (COTS) thermal IR camera with 50 mK noise-equivalent temperature difference (NEDT) with a single, broadband filter covering the IR wavelength region from 7 µm to about 14 µm, was adapted for measuring SO2 plumes. Detailed algorithms have been developed at NILU to calibrate and process the Cyclops imagery and create quantitative measures of SO2 emissions. Figure 1 shows some examples of plume stack SO2 measurements. The image data have been transformed into thermal temperature images and then to path concentrations (molecules cm-2) by using scene-based image analysis and radiative transfer theory.
Fig. 1 – Some examples of plume stack SO$_2$ measurements.

Fig. 2 – Volcanic SO$_2$ plumes from Villarica volcano, Chile, measured by a UV camera.

The NILU Ultra-violet imaging camera (NiluUVGasCam) is a scientific instrument for use in identifying and measuring polluting gases in the troposphere. The UV camera exploits a strong absorption feature of the SO$_2$ molecule between 280–320 nm. By calibrating the camera using gas cells containing known amounts of SO$_2$, the recorded light intensity can be related directly to the path concentration. Because the camera can sample rapidly (several images per second), features in the images can be tracked and the “in plume” wind speed and gas flux can be derived. False color ultraviolet images of the Villarrica (Chile) volcanic plume taken coincidentally from 16.5 km N of the volcano are shown in Fig. 2. The clean, cloud-free atmosphere provided an ideal background, and absorption of UV light by SO$_2$ in the volcanic plume allows distinct discrimination from the background sky.
The SO$_2$ plume demonstrates significant heterogeneity; calculated fluxes ranged over at least a factor of three in these plume images. This heterogeneity is difficult to resolve using a scanning technique [6].

The Data Center is in charge of processing, analyzing and storing the data collected from the ON and satellites, as well as for modeling, scenario building and forecast, using satellite imagery and modeling tools (FLEXPART, MAP3D) implemented with the help of Norwegian partner - NILU.

In order to establish source-receptor relationship for the measurements, the Lagrangian particle dispersion model **FLEXPART** http://transport.nilu.no/flexpart is used. FLEXPART has been validated with data from continental scale tracer experiments [7] and was used previously to study the transport of biomass burning and anthropogenic emissions between continents [8] and into the Arctic. The main purpose of FLEXPART model simulations is typically to identify the sources of the measured pollution and to quantify removal from the atmosphere occurring on the way. This enables a better interpretation of surface as well as vertical measurements (e.g. lidar).

**Satellite data** is also used in RADO. **EUMETCAST** is a system provided by ESA and industry to allow real-time reception of some important satellite data streams directly to the user. Our main region of interest is Romania and its surrounding countries and the coverage is extremely good for this system. Data streams include access to the primary European meteorological satellite – MSG, with its geostationary sensor, SEVIRI capable of providing data over Romania every 15 minutes. Methods are being developed to utilize satellite data with other measurements to probe deeper into the atmosphere and provide boundary layer estimates of gas amounts.

Urban pollution affecting human health is often concentrated in cities and the level of pollution is correlated with temperature. Hotter cities tend to have higher levels of pollution. Heat stress is also a key health hazard for older people and summer heat waves can put pressure on critical services, such as water and energy. Satellite data, because of its great spatial density and timeliness is very useful for monitoring and helping to forecast prolonged hot and dry spells in weather. Figure 3 shows some results of a study using MODIS satellite data (at 1 km spatial resolution) that illustrates quite well the urban heat island affect of Bucharest.

Land surface temperature (LST) is also important in climate studies and in numerical weather forecasting. New algorithms have been developed at NILU to determine LSTs over Europe for use in climatologically studies. An example of a monthly LST product for Europe is shown in Fig. 4. This technology will be part of the NILU EUMETCAST system and hence available for use in other applications for RADO.
Fig. 3 – Day and night satellite-derived surface temperatures for Bucharest showing the urban heat island effect.

Fig. 4 – Land surface temperatures determined from the (A)ATSR instrument and averaged over the month of August 2007.
Greenhouse and other polluting gases can be determined using satellite data. Perhaps of greatest interest is CO$_2$, but this gas has a very strong diurnal and seasonal cycle, is already abundant in the atmosphere and is difficult to measure from satellite. Nevertheless there are several instruments that have been able to retrieve mid-tropospheric CO$_2$ and a new instrument (OCO) is proposed for launch in 2009 that is wholly dedicated to measure the abundance of CO$_2$ in the atmosphere. SO$_2$ pollution sources exist within Romania and external to it, from neighboring countries. Some initial analyses of the OMI SO$_2$ (Fig. 5) data suggest that it is indeed measuring high levels of SO$_2$ trapped with the boundary layer. Other important atmospheric gases have been measured by satellite, including CO (Fig. 6), and NO$_2$ (Fig. 7).
3. RESULTS AND DISCUSSION

One of the most challenging tasks in RADO is to extract best information from available instrumentation. In situ monitors, such as nephelometers, gas analyzers, aerodynamic particle sizers and sun photometers, are also used as complementary techniques to lidars and modeling. Since each technique has
limitations in terms of dynamic range, accuracy and derived parameters, the information obtained is never complete and therefore subject of assumptions impossible to quantify. The vision of RADO is to put all the instruments together, to measure the same atmosphere and to correlate / combine information in order to deliver a much more complete picture of the reality. This is not always possible in a quantitative way, but qualitative analysis also give important results, such as inter-validation of various techniques, source identification, forecast, impact assessment.

This paper presents an example of data analysis and correlation at RADO, using lidar and nephelometer data in combination with air mass trajectory analysis. The monitoring of long range transported aerosols is regularly performed using a multiwavelength lidar system RALI which is part of EARLINET (European Aerosol Lidar NETwork) at Magurele site (44.35 N, 26.03E), in the SSW part of Bucharest. The laser radiation of RALI is emitted at 1064, 532 and 355 nm and collected at 1064, 532p, 532s, 355, 607, 387 and 408 nm. In order to increase the dynamic range, almost all channels have both analog and photon counting detection (analog for the lower troposphere and photon counting for the upper troposphere).

Data presented in this paper were recorded at 355, 532 and 1,064 nm. The laser beam was sent on the vertical up to 15 km altitude and the acquisition was set at 1 minute temporal resolution and 3.75 m spatial resolution. Calculation of aerosol backscatter coefficient at the 3 elastic wavelengths gives information not only about the layer altitude, but also on some aerosol characteristics such as aerosol probable class (urban, dust, smoke, maritime, mixed etc.) [12, 13]. Lidar data were compared/checked against corresponding nephelometer measurements, operating continuously. The system is part of Atmospheric Department Laboratory, Faculty of Physics and is located close to the multiwavelength lidar site. Nephelometer detects ground level aerosols and measures the properties of aerosol particles (total scattering and the backscattering coefficients) by detecting light scattered intensity at three wavelengths (450, 550 and 700 nm) using highly sensitive photomultiplier tubes. Every photomultiplier tube has a 40 nm bandpass filter. The data recorded have 1 minute temporal resolution.

The analysis of long-term statistics of nephelometer data evidenced a strange behavior between 18 and 19 July 2009 (Fig. 8a). Total backscatter at the 3 wavelengths (1 hour average) show a constant value until 18 July and a significant decrease starting from the morning of 18 July until the evening of the same day. The ratio of total forward and total backward scattering (Fig. 8b) also show a variation for the same period, by increased values compared with the mean of previous hours. In terms of color ratios, the values obtained decreased drastically from a mean and ordinary value of 0.8 to 0.5 (Fig. 8c).
Fig. 8a – Total backscatter coefficient at 450, 550 and 700 nm derived from nephelometer.

Fig. 8b – Total forward per backscatter coefficient at 450, 550 and 700 nm derived from nephelometer.

Fig. 8c – Color ratios at 550/450 and 700/550 nm derived from nephelometer.
First information that can be extracted refers to a mixing of common local aerosols with particulates having different optical characteristics. The increase of forward-to-backward scattering indicates particles with higher asphericity than the common local aerosols, while the decrease of color ratios indicates smaller particles. This behavior could be explained by a local pollution event or by a mixing of long-range transported aerosols and local ones during the nighttime syncopation of the Planetary Boundary Layer.

In order to exclude one or the other, lidar vertical profiles were analyzed for the same period. Range Corrected Signal time series and layer height retrieval for 18 (Fig. 9a) and 19 July (Fig. 9b) show the presence of an approximate uniform structure of layers up to 3.5 km altitude on 18 July which evolves in a more distinct layer structure on 19 July. The atmosphere near the ground is not visible due to the “blind” region of the lidar; therefore no low-level pollution event could be evidenced by lidar. Still, it can be noted from the RCS on 18 the descending trend of the PBL after sunset, acting as a piston on low-level aerosols and leaving an open gate to the aerosols in the Free Troposphere.

Fig. 9 – Range Corrected Signal time series and layer top retrieved from lidar on:
Optical characteristics of the aerosols in the 2 to 3.5 km altitude range were assessed using the multiwavelength lidar data. We obtained for both datasets a particle depolarization coefficient of 1.9% for the 3.5 km layer, which suggest a non-spherical shape. Moreover, color ratios at 532/355 nm having values of approximate 0.6±0.1 and Angstrom coefficients at 355/532 nm of 0.3±0.1 suggested a significant contribution of fine mode to the particles size distribution.

To identify the origin of air masses reaching our site at those altitudes, the version 4 of the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) [14] was used. As inputs of the model we used the coordinates of lidar location and specific levels at 1,000 m, 3,000 m and 5,000 m altitude. These values were chosen based on the backscattering coefficient profiles previously derived. The model was run backward for 2 days and backward trajectories are shown in Fig. 10a. The model showed that air masses arriving at 1 and 3 km altitudes came from Ukraine, where lots of forest fire was signalized by MODIS during that week (Fig. 10b).

Biomass burning aerosol consists of two major chemical components: black carbon (BC), which primarily absorbs solar radiation, and organic carbon (OC), which primarily scatters solar radiation. The intrusion of smoke-like aerosols coming from Ukraine should also determine an increase of the extinction, since the major component of these particles is strongly absorbing light. Therefore, to confirm our conclusion we also investigate AERONET retrievals for that period.

AERONET (AErosol RObotic NETwork) is a world-wide network of automated ground-based CIMEL sunphotometers providing spectral aerosol optical depth (AOD), inversion products of other aerosol optical properties, such as single scattering albedo (SSA) and the column integrated aerosol size distributions above the measurement site [15].

Data provided by AERONET based on sun photometer measurements at our site show an increase of the AOD for the same period (Fig. 11a). Level 2 products from AERONET confirmed aerosols type as “biomass burning”, based on specific size distribution having the fine mode well represented (Fig. 11b).

Due to the fact that the nephelometer measures continuously, the simple analysis of long-term statistics can evidence special events to be further investigate by lidar and back-trajectories. Although lidars and nephelometer cannot give comparable parameters due to specific limitations of each technique, cross-analysis of their data can be used for quick assessment of aerosol properties, type and origin.

For the case study on 18-19 July 2009, data recorded by the nephelometer showed increased values of total backscatter coefficient, stronger forward-to backward scattering coefficient and lower color ratios. The conclusion was that smoke-like aerosols coming either from local source or from long-range transport penetrated the low-level atmosphere, inducing significant variations of optical properties. During same days lidar measurements evidenced the presence of free troposphere aerosol layers between 1,700 and 3,500m altitude. Further analysis
based on back-trajectories and sun photometer retrievals proved that, although in general uplifted particles have a negligible influence at ground, in certain meteorological conditions, they can reach the surface and can be detected by ground-based in situ instruments.

Fig. 10a – Backward air mass trajectory for 3 days starting July 18, 20:00 UTC.

Fig. 10b – Biomass burning, MODIS July 18, 2009.
Fig. 11a – Aerosol Optical Depth derived from sun photometer for July 2009.

Fig. 11b – Aerosol size distribution derived from sun photometer data on July 18, 2009.

4. CONCLUSIONS

The combination of lidar observations with in situ measurements and models provides a unique opportunity to conduct long-term inter-calibrations and complementary or simultaneous monitoring of different atmospheric parameters over various space-time scales at the Romanian Atmospheric 3D research Observatory – RADO.
Main function of the observatory is to perform experimental and theoretical research for atmospheric composition and air quality assessment, including impact on climate and climate variability. Observational data (air chemistry, layering, meteorology) are collected at 4 urban sites (Baneasa – North Bucharest, Iasi, Timisoara and Cluj-Napoca) and one super site (Magurele – South Bucharest).

Beyond operational monitoring activities, advanced data processing, analysis and correlations is performed to study the planetary boundary layer, climatology of aerosols, their impact on cloud formation, correlation process involving various pollutants, seasonal and regional characteristics.

This paper presents an example of data analysis and correlation at RADO, using lidar and nephelometer data in combination with air mass trajectory analysis.

Optical characteristics of the aerosols in the 2 to 3.5 km altitude range were assessed using the multiwavelength lidar data. The conclusion was that smoke-like aerosols coming either from local source or from long-range transport penetrated the low-level atmosphere, inducing significant variations of optical properties for local aerosols. Source identification was done based on layer altitudes from lidar and air mass backtrajectory analysis.

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