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New atmospheric composition observations in the Karakorum region: Influence of local emissions and large-scale circulation during a summer field campaign

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HIGHLIGHTS

- First use of a new transportable and embedded measurement station.
- First indication about summer aerosol, O_3 and CO_2 variability in the Karakorum.
- Domestic combustion appears as a possible source of high aerosol concentration.
- Thermal circulation dominated the diurnal variability of O_3, CO_2 and particles.
- Ozone, CO_2 and aerosol day-to-day variations were linked to large scale circulation.

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ABSTRACT

In this work we provide an overview of short lived climate forcers (SLCFs) and carbon dioxide variability in the Karakorum, by presenting results deriving from a field campaign carried out at Askole (3015 m a.s.l., Pakistan Northern Areas), by Baltoro glacier. By using an innovative embedded and transportable system, continuous measurements of aerosol particle number concentration (Np, 1571 ± 2670 cm^{-3}), surface ozone (O_3, 31.7 ± 10.4 nmol/mol), carbon dioxide (CO_2, 394.3 ± 6.9 μmol/mol) and meteorological parameters have been performed from August 20th to November 10th 2012. The domestic combustion from the Askole village emerged as a possible systematic source of contamination in the valley, with short-lasting pollution events probably related to domestic cooking activities characterized by high values of Np (6066 ± 5903 cm^{-3}). By excluding these local contamination events, mountain thermal wind regime dominated the diurnal variability of Np, O_3 and CO_2. In comparison to night-time, we observed higher Np (+354 cm^{-3}) and O_3 (+7 nmol/mol) but lower CO_2 (−8 μmol/mol) in air-masses coming from the lower valley during the central part of the day. Part of the day-to-day atmospheric composition variability can be also ascribed to synoptic circulation variability, as observed by using HYSPLIT 5-day back-trajectories.

1. Introduction

Karakorum is home to hundreds of peaks which are higher than 6000 m above sea level (a.s.l.), and it bears extensive mountain glaciers (e.g. Young and Hewitt, 1990). Currently, this mountain range is widely investigated because of the so-called "Karakorum anomaly" (Hewitt, 2005), i.e. a general stability of many central Karakorum glaciers versus the general shrinking of eastern Himalayan and Tibetan glaciers (e.g. Kehrwald et al., 2008; Gardelle et al., 2012).

Recently, field experiments, in situ observations, and satellite monitoring in various regions of the world have pointed to the existence of wide polluted tropospheric layers (the so-called

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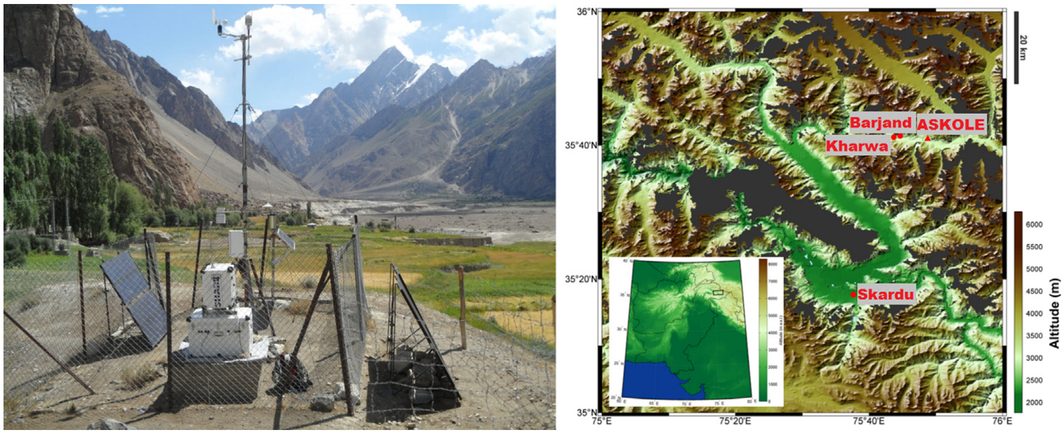

Fig. 1. On the left, the embedded station at Askole. On the right, a geographic map of the Braldu valley with Askole position, as well as other villages. The insert denotes the Braldu valley position with respect to Karakorum region.

Atmospheric Brown Clouds – ABC) which typically consist of atmospheric aerosols and reactive gases and their implications for global climate were rapidly recognized (Ramanathan and Carmichael, 2008). In South Asia, the ABC phenomenon (Ramanathan et al., 2007) has important regional climate impacts, with strong perturbations of the regional radiative balance both at the surface and within the atmosphere, and with strong impacts on the hydrological cycle and monsoon regimes. Furthermore, the South Asian ABC is potentially responsible for a reduction of crop production and changes in snow cover dynamics in the Hindu-Kush-Himalayan-Tibetan area, with possible consequences for the freshwater supply over Southern and Eastern Asia and for agricultural purposes.

Currently no systematic information about aerosol and trace gas levels and variability exists on the Karakorum region, where logistical difficulties and adverse meteorological conditions have often limited observations.

The purpose of the present study is to provide information on atmospheric variability from an area where no information exists. A new embedded-transportable monitoring system, specifically designed to perform atmospheric observations in remote regions, has been deployed to this aim at the village of Askole (3015 m a.s.l., Pakistan Northern Areas) from August 20th to November 10th 2012. With this original miniaturized observing station, we documented aerosol number concentration and surface O_3 and CO_2 mixing ratios in the Karakorum region, also investigating the influence of local emissions and long-range transport.

2. Experimental and methods

2.1. Measurement site

Atmospheric and meteorological observations have been performed 300 m from the Askole village (35°40′50″N, 75°48′55″E, ca. 600 inhabitants), the last village on the route to the Baltoro Glacier (60 km long and with an extension of approximately 700 km^2, it is considered one of the largest valley glaciers in the world), at an altitude of 3015 m a.s.l. (Fig. 1). The village is on the right side of the Braldu valley (oriented W–E). The whole installation was protected by fencing and it was located in the middle of a crop area. The only source of pollution at Askole is represented by domestic (heating and cooking) and agriculture (fire of crop vast) biomass burning, since no traffic emissions exist. Along the valley, several villages are present: Korphe (located ca. 1 km away), Breadang (3 km), Barjand and Kharwa (6 and 8 km, respectively). The major cities are Skardu (ca. 30,000 inhabitants), located 46 km from the measurement site at the confluence of Indus and Shigar rivers and Gilgit (ca. 216,000 inhabitants, at 140 km). The choice of the Askole location was related to the necessity of obtaining information about the variability of atmospheric composition by the Baltoro glacier, with a particular emphasis on the possible role of the along-valley thermal circulation. The proximity of the village, as well as the installation inside a fenced experimental field where an automated weather station (AWS) with its solar panels is already located, ensured the instrument safety and supply of electrical power.

During summer months, meteorological conditions at Askole are predominantly dry and precipitations related to the wet monsoon are episodic and scarce, as also stated by Palazzi et al. (2013) (see Table 1). Wind speed (WS) over the whole campaign showed a rather low average value (2.2 ± 1.4 m/s). The local wind is strongly affected by the development of thermal circulation along the valley, with westerly up-valley winds during the day and nighttime easterly down-valley winds, as a consequence the wind direction (WD) showed a bidirectional distribution, with dominating directions from East and West of the valley. The typical diurnal variation of the wind is characterised by higher WS (>2.5 m/s) coming from SW in the central part of the day, and weaker WS (1.3–2.5 m/s) coming from E–NE during the remaining part of the day (Fig. 2).

2.2. Instrumental setup

The trace gases (surface O_3 and CO_2) and aerosol measurements have been performed using a newly developed compact embedded station. The miniaturized station has the following technical characteristics:

- The system is equipped with an integrated weather station (WXT520, Vaisala), a condensation particle counter (CPC, 3010,

Table 1
Main statistic values of the meteorological parameters collected at Askole over the whole field campaign.

Parameter	Average	St. Dev.	Max	Min	25th perc.	75th perc.
Wind speed (m/s)	2.2	1.4	12.0	0.1	1.2	2.9
Air temperature (°C)	10.2	5.9	32.4	−2.5	6.0	13.6
Relative humidity (%)	43.2	19.4	89.4	9.5	27.5	57.6
Rain precipitation (mm)	0.6	1.2	14.2	0.0	0.0	0.3
Air pressure (hPa)	707.7	2.6	714.4	700.4	705.9	709.5

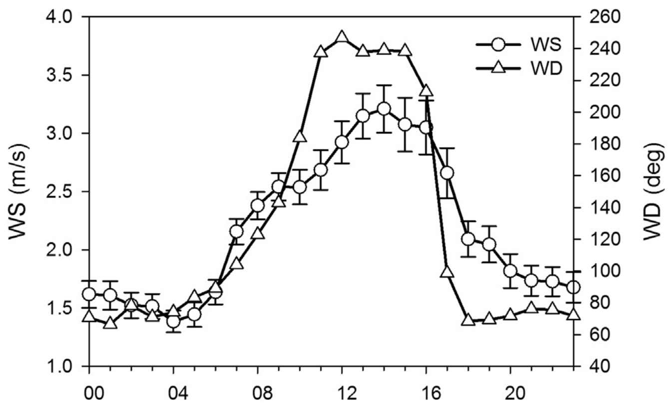

Fig. 2. Average diurnal variations wind speed (WS) and direction (WD). For WS, the vertical bars denote the expanded uncertainties ($p < 0.05$) of the mean.

TSI) which provides total number concentration for particles with diameters 15 nm < Dp < 3 μm (hereinafter referred to as Np), an O_3 analyser (Ozone Monitor 202, 2B Technologies) and a probe for CO_2 measurements (CARBOCAP GMP343, Vaisala).

- The system is made up of two different parts that can be easily assembled. The first part (38 × 50 × 65 cm and 18.5 kg approximately) contains all the instrumentation, while the second (80 × 60 × 40 cm, and 80 kg) is used for storage of standards, chemicals and batteries. Power consumption is limited (50 W), which allows the system to be powered by solar panels.

For a more detailed description about the instrumental setup and the intercomparison test performed at Mt. Cimone station (Italy), please see Section S1 of the supplementary material.

Due to the harsh experimental conditions, the data coverage varied between different instruments. Meteorological parameters and CO_2 data have been collected for the whole campaign period (with a major data gap on October 19th–31st due to a failure of the acquisition system), while O_3 and Np were collected until October 7th and September 23rd, respectively. In this work, time is expressed as local time (UTC+4), surface O_3 and CO_2 are expressed as STP conditions, while aerosol concentration by the CPC is expressed as ambient concentration.

2.3. HYSPLIT back-trajectories

In order to determine the synoptic origin of air-masses reaching the measurement site, 5-day back-trajectories were calculated every 6 h (at 4:00, 10:00, 16:00 and 22:00) with the HYSPLIT back-trajectories model (Draxler and Hess, 1998). The model calculations were based on the GDAS meteorological field produced by NCEP with a horizontal resolution of 1° × 1°. Sub-grid scale processes, such as convection and turbulent diffusion, were not represented by the model. Back-trajectory ensembles were calculated, with endpoints shifted by ±1° in latitude/longitude to partially compensate such uncertainties. The use of back-trajectories based on coarse meteorological input data could be very challenging in regions characterised by complex mountainous topography. For this reason, a sensitivity study has been conducted to evaluate the possible impact of the model topography to the back-trajectory calculation: several model runs have been performed by slightly changing the altitude of the starting points of the back-trajectory within a vertical range from 100 to 2500 m a.g.l. This experiment resulted in a nearly identical outcome for each set of trajectories, so that more confidence in the results was obtained.

In order to identify the main flows of the synoptic-scale occurring at the measurement site, a cluster analysis (Draxler, 1999) was applied to the HYSPLIT back-trajectory ensembles. The clustering was applied to all single elements of each back-trajectory ensemble. At each step of the process, the appropriate number of clusters was determined by observing the variation of several statistical parameters (i.e. total trajectory dispersion and total root mean square deviation). Cluster analysis is a multivariate statistical technique that can explore structures given a dataset (for basic details see: Anderberg, 1973; Everitt, 1980). A number of different clustering methodologies were used in the past to investigate the variability of atmospheric composition as a function of atmospheric circulation (e.g. Dorling et al., 1992; Jorba et al., 2004 and references therein; Hondula et al., 2010 and references therein). Basically, by maximizing between-group variance and minimizing within-group variance, these methodologies can identify similar air-mass (back-) trajectories and aggregate them. However, it should be noted that even if the cluster analysis could remove some subjectivity in the aggregation processes, nevertheless this is far to be a true objective method for classification (especially due to the choice of the clustering algorithm, the specifications of the distance measure, and the number of optimal clusters, see Stohl, 1998; Jorba et al., 2004). Moreover, as reported in Mace et al., 2011, further uncertainties can arise from steep emission gradients along similar shaped trajectory clusters.

3. Results and discussion

3.1. Synoptic-scale air-mass circulation

The analysis of back-trajectory clusters led to the identification of 6 main patterns for synoptic-scale atmospheric circulation affecting the Northern Areas of Pakistan during the experimental campaign: WES-Westerly, TAK-Taklamakan, LOC-Local, SOU-Southern Pakistan, INS-Indian Subcontinent and NOR-Northerly (Fig. 3). Westerly circulation (WES) prevailed for 26.2% of back-trajectories, with 5-day old air-masses originating in the region

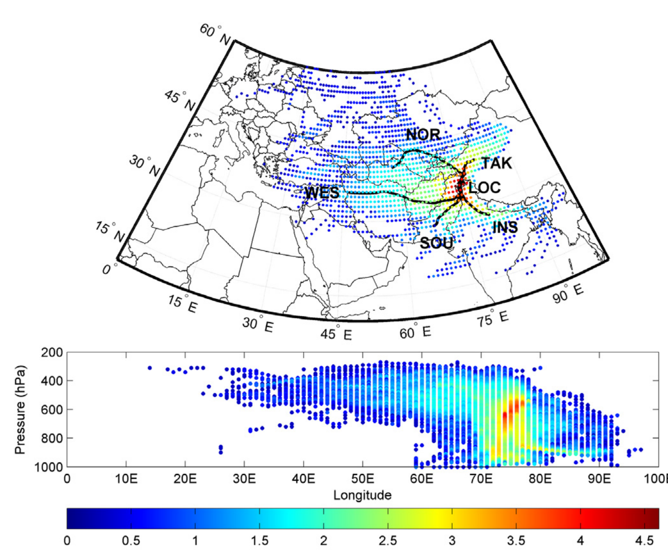

Fig. 3. Concentration field (vertical cross section included) for HYSPLIT back-trajectory points ending at Askole and centroids of back-trajectory clusters (WES-Westerly; TAK-Taklamakan; LOC-Local; SOU-Southern Pakistan; INS-Indian Subcontinent; NOR-Northerly). Only back-trajectories which did not come in contact with surface terrain in the last 24 h before reaching the measurement site have been considered.

extending from the Mediterranean basin and Afghanistan, which rarely travelled above the 800 hPa pressure level. A not negligible fraction of air-masses also originated or travelled over the Taklamakan desert (TAK: 16.2%), at altitudes ranging from 800 to 300 hPa. “Regional” air-masses originated or recirculated over the northern (LOC: 19.1% of occurrences) or southern Pakistan (SOU: 8.0%). 23.8% of the air-mass occurrences were related to the INS back-trajectory cluster that represents the typical circulation affected by the summer monsoon system. A small occurrence of northerly air-masses (NOR) has been also detected (6.7%): they were characterised by relatively high travel altitudes (typically 700 hPa, rarely moved below 600 hPa before reaching the measurement site) and originated in a region that spans from eastern Europe to central Asia.

3.2. Variability of atmospheric composition

3.2.1. Local contamination events

Fig. 4 reports the time series of CPC particle concentration (Np, average value $\pm 1\sigma$: 1571 $\pm$ 2670 cm^{-3}, $N = 2864$; median: 756 cm^{-3}), O_3 (31.7 $\pm$ 10.4 nmol/mol, 3711; 32.9 nmol/mol) and CO_2 (394.3 $\pm$ 6.9 µmol/mol, 6057; 393.3 µmol/mol).

The most striking feature in the Np time series is the presence of systematic concentration peaks which occurred at the early morning (from 4:00 to 7:00) and in the evening (from 16:00 to 20:00), as also pointed out by the analysis of the average diurnal variations (Fig. 5).

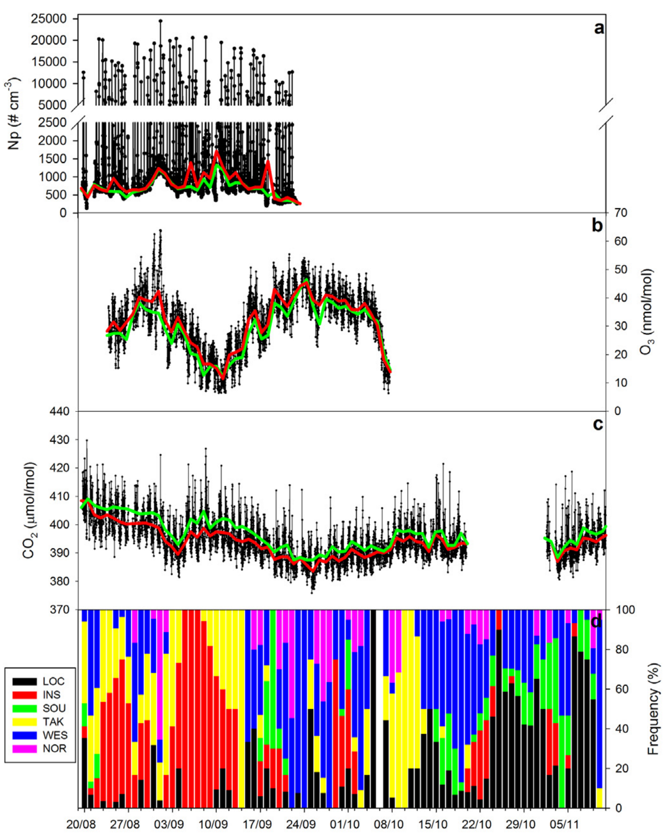

Fig. 4. Time series of CPC particle number concentration (Np, panel a), surface ozone (O_3, panel b), carbon dioxide (CO_2, panel c) and daily air-mass circulation occurrences (panel d, WES-Westerly; TAK-Taklamakan; LOC-Local; SOU-Southern Pakistan; INS-Indian Subcontinent; NOR-Northerly). Red lines denote daily averages (computed excluding the “local contamination events”), while green lines represent night-time (between 21:00 and 4:00) averages.

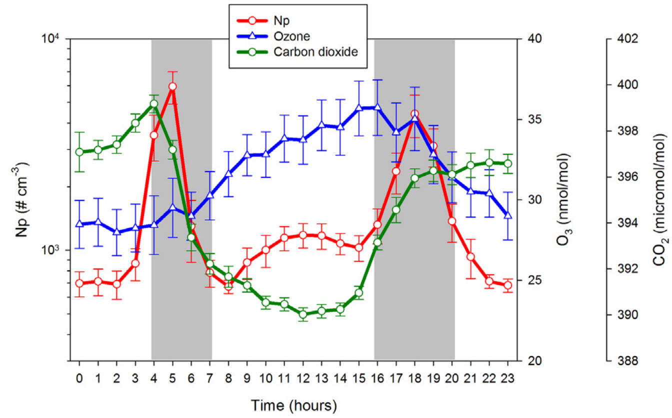

Fig. 5. Typical diurnal variations for Np (red), O_3 (blue) and CO_2 (green). The vertical bars denote the expanded uncertainties ($p < 0.05$) of the mean, while the shadow areas indicate the periods possibly affected by the “local contamination events”.

A detailed inspection of the internal working parameters of the embedded station did not reveal any evident malfunctioning of the system which could explain these anomalous peaks. The time of day at which these peaks occurred is almost simultaneous to the time at which the people from the village burn wood and other organic material, especially for cooking. At Askole, during the summer season, the population uses very simple cooking systems, which are often represented by open fires with rough chimneys that guarantee the smoke ventilation from traditional houses to the outdoor. The fuel typically consists of wood that people collect in the neighbourhood. It is thus conceivable that the particle peaks could be ascribed to the domestic emissions from the near village. During these events, with respect to other measurement periods, the wind direction was dominated by north-easterly components (not shown here), thus providing a confirmation of possible smoke transport from the village. The absence of atmospheric mixing in the morning hours is supported also by the analysis of the wind (Fig. 2): as already introduced in Section 2.1, WS shows very low values during the morning hours (reaching a minimum between 4:00 and 5:00), while it increases during the day, simultaneously with a change in WD. It then decreases after 17:00, although WS values during the evening are little higher than the morning minimum.

We selected as “local contamination events” the measurement periods 4:00–7:00 and 16:00–20:00 (when the Np peaks were observed) and the periods for which the condition $Np(t) - Np(t-1) > Np\ (t-1)*0.5$ is verified, where $Np(t)$ represents the total particle number concentration at time t. The methodology for selection that we applied is based on the definition of appropriate time windows, as well as on the analysis of Np particle behaviours, in order to evaluate the frequency of occurrence of these polluted events. The local contamination events excluded by this condition were characterized by abrupt but short-lasting (less than 2 h) increases of aerosol particle number. By applying this selection, 31% of measurement period has been identified as influenced by local pollution (27% of CO_2 and 26% of O_3 data). In comparison with the remaining data, CO_2 (a well-known tracer for combustion processes) was characterised by a limited increase (+2 µmol/mol) during these periods. Nevertheless, during these episodes the highest CO_2 mixing ratios were usually observed at Askole. On average, no statistically significant variations of O_3 were observed during these contamination events.

3.2.2. Influence of the wind circulation across the valley

The mountain wind regime (as reported in Section 2.1) clearly influenced the diurnal behaviour of atmospheric composition at the measurement site. Indeed, aerosol particles and trace gases showed a typical diurnal cycle. In particular, the lowest particle concentrations were observed at night (from 22:00 to 02:00, mean value: 686 cm^{-3}) while a statistically significant (99% confidence level) increase took place during day-time (from 11:00 to 15:00, mean value: 1056 cm^{-3}). This behaviour suggested that aerosol particles from the lower troposphere were transported to Askole by up-valley thermal circulation, while during the night cleaner air-masses, which are more representative of the upper troposphere, were transported by down-valley winds. Results obtained at Askole can be compared with results observed at NCO-P, in the central Himalayas (see Fig. S1 in the auxiliary material). Concerning Np diurnal variability, night-time values were similar at both sites, while a large Np peak affected NCO-P during the central part of the day, which is attributed to the occurrence of systematic nucleation events at the Himalayan station (Venzac et al., 2008; Sellegri et al., 2010); thus nucleation events do not seem to be one of the main mechanisms influencing particle number at Askole, which are on the other hand mainly affected by valley wind transport.

As pointed out by this analysis, the surface O_3 average diurnal variation was also characterized by lower values during the night (00:00–4:00, mean value: 28.5 nmol/mol) and a peak during the afternoon (14:00–15:00, mean value: 34.6 nmol/mol), with average diurnal cycle amplitude of 6.1 nmol/mol. On average, the diurnal O_3 peak occurred a few hours later compared to the Np peak and the maximum of the up-valley wind speed. As also reported from other mountain stations around the world (see e.g. Cristofanelli et al., 2010; Cristofanelli and Bonasoni, 2009; Fischer et al., 2003), air-masses coming from the valley were richer in O_3 possibly due to the photochemical production related to the regional-scale anthropogenic precursor emissions. This is different from the simultaneous observations at NCO-P (see Fig. S2 in the auxiliary material) which were characterized by a smaller and earlier O_3 increase (+4 nmol/mol) which is typically observed from the morning hours until noon, in agreement with Cristofanelli et al. (2010). The presence during day-time (nigh-time) of air-masses representative of the lower troposphere (free troposphere) is also testified by the CO_2 average diurnal variation, which showed lower mixing ratios (on average: −8 μmol/mol) in respect to night-time values. Indeed, during summer season, air-masses from the atmospheric boundary layer are depleted in CO_2 in respect to free-troposphere or upper tropospheric air-masses due to the vegetation uptake by photosynthesis (e.g. Colombo et al., 2000). As reported by Colombo et al. (2000), mixing ratio in summer northern Hemisphere is higher in the middle/upper troposphere than in the lower troposphere, since the diurnal vegetation activity at the ground produces a decrease of CO_2 which is slowly propagated to the upper atmospheric layers. In agreement with observations in Nepali Himalayas (e.g. Venzac et al., 2008; Bonasoni et al., 2010) these results clearly suggested that the Braldu valley can represent a channel throughout which anthropogenic pollutants like aerosol particles and surface O_3 can be transported up to the high Karakorum. As often observed, thermal circulation is one key process driving their variability in mountain areas.

3.2.3. Comparison with other high altitude sites and influence of large-scale circulation

Excluding the "local contamination events", the Np median concentration at Askole (676 cm^{-3}) is comparable with the median value (657 cm^{-3}) obtained during the same period at the NCO-P. For NCO-P the Np variability appeared to be in agreement with early measurement results provided by Sellegri et al. (2010). However, it should be pointed out that NCO-P is strongly affected by the occurrence of new particle formation events, which can significantly affect Np. With the aim of comparing Askole results with other mountain measurements performed in Asia, we calculated Np for STP conditions (i.e. 1013 hPa and 0 °C). At the Mukteshwar station (at the foothills of the Indian Himalayas at 2180 m a.s.l.) and at Mount Waliguan (China, 3816 m a.s.l.), as presented by Komppula et al. (2009) and Kivekäs et al. (2009), average particle (10–800 nm) number concentrations were more than 50% higher (3480 cm^{-3} and 3280 cm^{-3}, respectively) than the ones collected at Askole (1038 cm^{-3}). The Mukteshwar station, located at a lower height, is much more influenced by the very high concentrations encountered in the boundary layer of the Indo-Gangetic plains.

By neglecting the "local contamination events", O_3 average values (31.8 ± 9.9 nmol/mol) were lower than the mixing ratios observed during the same period at the NCO-P (38.6 ± 8.0 nmol/mol), see also Fig. S2 in the supplementary material. The average O_3 mixing ratios observed at the NCO-P, were not statistically different from the "representative" monsoon values reported by Cristofanelli et al. (2010) for years 2006 and 2007 (39 ± 10 nmol/mol). The lower O_3 values observed at Askole were influenced by the low O_3 period (with mixing ratio ranging from 10 nmol/mol to 20 nmol/mol), observed from September 7th to 12th and October 6th to 9th 2012 (Fig. 4(b)), when INS and TAK air-masses were present at the measurement site according to HYSPLIT back-trajectories (Fig. 4(d)). These values seem to be comparable with those observed at Mt. Abu (24.6°N, 72.7°E, 1680 m a.s.l., India) and at Nainital (29.37°N, 79.45°E, 1958 m a.s.l.), during southwest monsoon period (monthly average about 25 nmol/mol), when clean marine air from the Arabian Sea and Indian Ocean influence these measurement sites (Naja et al., 2003; Kumar et al., 2010). As pointed out by laboratory/model studies (e.g. Hanisch and Crowley, 2003; Bauer et al., 2004) and atmospheric observations (e.g. Bonasoni et al., 2004; Umann et al., 2005), mineral dust may decrease tropospheric O_3 due to heterogeneous chemistry and a decreased efficiency of photochemical production. It is thus conceivable that air-masses coming from Taklamakan desert were characterised by low O_3 values.

During these periods, increases of CO_2 daily values have also been observed at Askole. This further supports the presence of both INS and TAK air-masses at the measurement site. Indeed, due to the stronger anthropogenic emissions, CO_2 is expected to have high source over the Indo-Gangetic plains (e.g. Baker et al., 2010); while the Taklamakan desert, having an arid climate and being covered by barren land or sparse vegetation, can be considered as a region poor of sinks of CO_2 (Hou et al., 2013), especially during the vegetative season. As reported by WMO (2013), the mole fractions of CO_2 in northern low latitudes (30°N − EQ) lags behind than in high latitudes by one or two months, resulting in an annual cycle with a maximum around May and minimum values in September. This feature is visible also from our observations, which are representative of the minimum phase of annual CO_2 cycle: despite the day-to-day variability, Askole showed the lowest CO_2 values at the end of September with increasing values moving throughout October.

As already mentioned, besides being affected by a significant diurnal variability, the atmospheric composition measurements at Askole were also characterised by significant day-to-day variations (Fig. 4), thus indicating that processes occurring at synoptic (or larger) scales can play a role in determining aerosol and trace gas behaviours in this remote region. With the aim of specifically investigating this point, according to Chevalier et al. (2007), we calculated the daily variability/hourly variability ratio for the observed SLCFs and CO_2. The ratio is in the range 80–90% for O_3 and CO_2, but drops to 60% for Np. It means that, in respect to day-to-day changes, processes occurring at diurnal time scale take only a minor

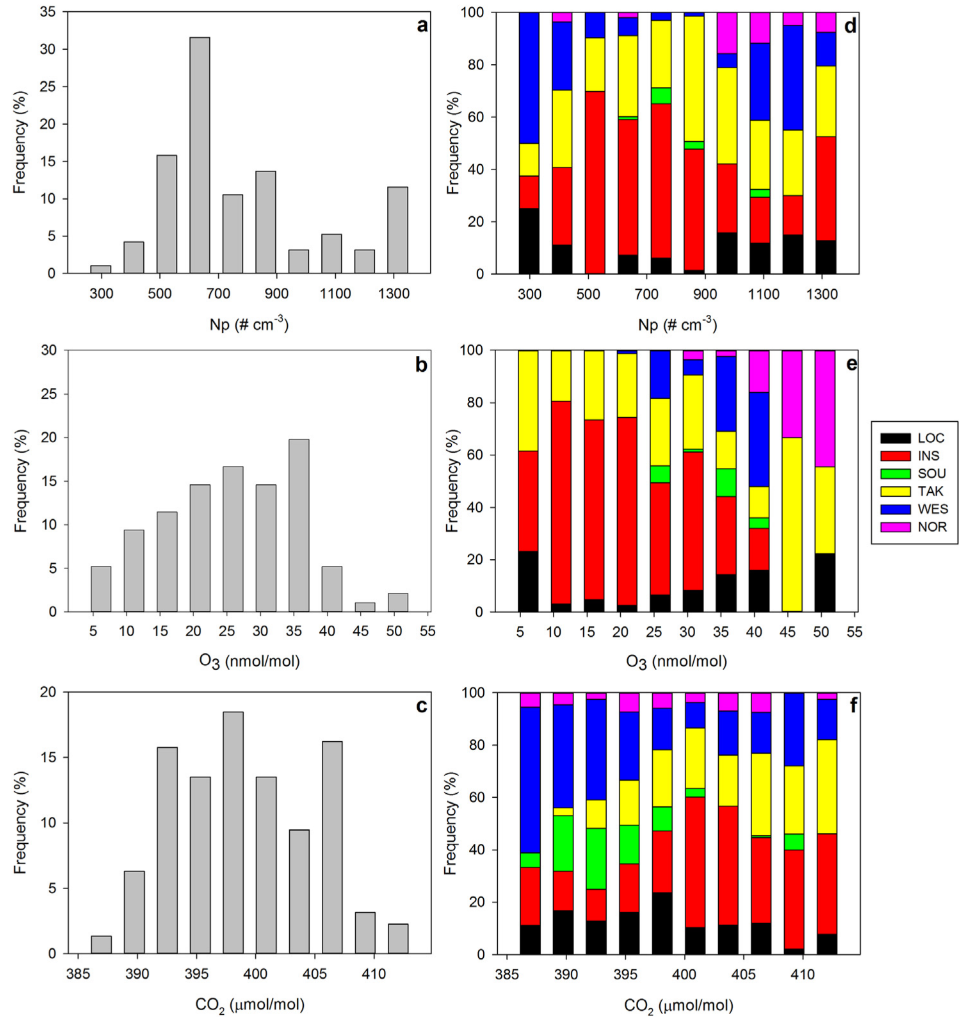

Fig. 6. Statistical distribution of nigh-time hourly Np (a), O_3 (b) and CO_2 (c) values at Askole during the field campaign. Percentage contribution of air-mass circulation occurrence (LOC-Local; INS-Indian Subcontinent; SOU-Southern Pakistan; TAK-Taklamakan; WES-Westerly; NOR-Northerly) for each bin of the distributions are reported (d–f).

part, even if not negligible (10–20%), in determining the total variability of O_3 and CO_2, but a considerable part (more than one third) for fine particle concentration.

For a systematic investigation of the possible relationship of synoptic air-mass circulation and atmospheric composition at Askole, hourly Np, O_3 and CO_2 values were analysed as a function of the different air-mass clusters. In order to capture the dominant synoptic-scale circulations and minimize the interference of the regional atmospheric boundary layer, only nigh-time data (less influenced by the development of daytime upward thermal transport processes occurring at mesoscale), which were not influenced by the local contamination events (see Section 3.2.1) were considered for this analysis. For night-time period (between 21:00 and 4:00) the following average values have been obtained: Np (average $\pm 1\sigma$: 681 ± 363 cm^{-3}, $N = 888$, median 676 cm^{-3}), O_3 (29.4 ± 9.6 nmol/mol, $N = 1176$) and CO_2 (396.8 ± 6.8 μmol/mol, $N = 1973$). Moreover, we only took into consideration the data for which at least 4/9 members of the back-trajectory ensemble did not come in contact with the surface terrain in the last 24 h before reaching the measurement site, with the aim of providing a robust relationship between synoptic-scale air-mass circulation and atmospheric composition variability.

Fig. 6 (panels a–c) reports the statistical distribution of the hourly Np, O_3, and CO_2 values for their whole measurement period (i.e. 35 days for Np, 45 days for O_3 and 72 days for CO_2). We also calculated the same distribution for the period of all the measurements simultaneously available (not shown here) and no significant differences have been obtained. For each observed parameter and distribution bin, we also calculated the relative percentage of the air-mass cluster occurrence (Fig. 6, panels d–f).

Considering Np, it is difficult to find a clear relationship with air-mass circulation clusters. However, it should be noted that the peak of the frequency distribution (600 cm^{-3}) is related to “long-range” circulation (i.e. INS and TAK). INS contributions appeared to be the dominant one for “intermediate” Np values (from 500 to 900 cm^{-3}). This can be understood as indicative particles concentration background in the free troposphere over the HKKH region, driven by long-range transport. Long-range transport includes contribution from TAK, possibly pointing out the role of mineral dust transport in the fine particle range, although this cannot be proven by our instrumental setup. Nevertheless, the possible presence of air-masses which are richer in mineral dust from Taklamakan desert was supported by the analysis of the OMI UV-Aerosol Index (see Section S2, supplementary material), which showed increased values over Askole, together with the presence of TAK air-masses at the measurement site (as deduced by HYSPLIT back-trajectories). In parallel, INS air-masses also contribute for the 50% of the upper Np values (>1300 cm^{-3}), indicating that long-range transport from South Asia is a major source of fine particles in the Karakorum.

As deduced by this analysis, the contribution of TAK and INS air-masses is dominant for low O_3 mixing ratios (<20 nmol/mol), but still significant up to 30 nmol/mol. As already discussed, this could indicate a possible influence of air-masses which originated from the Taklamakan desert or related to the summer monsoon circulation in determining the lowest values of the distribution peak. As shown in Fig. 6(d), a generally low occurrence of WES and NOR circulation was observed for the different O_3 values. Only the upper O_3 values (higher than 35 nmol/mol) showed not negligible contribution related to WES and NOR circulations, suggesting that O_3-enriched air-masses from the free troposphere could contribute to the episodic occurrence of these higher O_3 levels at the measurement site. With the aim of specifically discussing the possibility that stratospheric air-mass intrusions could affect Askole, we analysed the travel altitude and paths of these trajectories, as well as the spatial distribution of the total column O_3 (TCO), retrieved by OMI satellite instrument (product OMTO3d.003, see Levelt et al., 2006) and the in situ behaviour of RH (Bracci et al., 2012). 21.4% of days with NOR back-trajectories were characterized by air-masses which were travelling at altitudes that were higher than 300–400 hPa, and crossing areas characterized by steep gradients of the TCO field. During these days, the occurrence of low RH values (down to 11%, hourly average) further supported the possible occurrence of stratospheric intrusions (Stohl et al., 2003; Bracci et al., 2012).

The relationship between air-masses circulation appeared to be almost “reversed” for CO_2. Indeed, the lowest CO_2 mixing ratios were mostly associated with WES circulation (with smaller contributions from LOC, INS and SOU), while the highest values were mostly observed together with INS and TAK circulations.

4. Summary and conclusions

During summer 2012, an intensive experimental campaign has been carried out at Askole (3015 m a.s.l., Pakistan Northern Areas) to provide a first indication about the variability of atmospheric composition and average levels in the Karakorum. To our knowledge, these measurements are unique in this area of the world.

Measurements suggested that domestic combustion from the nearby village of Askole could represent a possible source of short-lasting pollution events with high aerosol concentration. Due to the presence of a thermal wind regime along the Braldu valley, where Askole is located, these aerosols have been rapidly dispersed along the valley. Outside of the period contaminated by local emissions, measurements were clearly representative of the variability at regional scale, with night-time and day-time concentrations close to those observed in the central Himalayas at the Nepal Climate Observatory – Pyramid (5079 m a.s.l.) and ranging from 500 to 800 cm^{-3}.

Mountain thermal wind regime also affects the diurnal variability of surface O_3 and CO_2. In particular, air-masses coming from the lower valley enriched in photochemically-produced O_3 (+6 nmol/mol compared to night-time) were systematically observed at Askole. Assuming that the wind speed typically observed at Askole during day-time (from 2.5 to 3.5 m/s) could be representative of the circulation across the valley, the influence of regional-scale emissions appears to be likely.

Lastly, by means of 5-day air-mass back-trajectory cluster analysis, possible influence of synoptic scale circulation has been assessed. Starting from the methodology proposed by Rozwadowska et al. (2010), we assessed that the variability of 5-day old air-mass clusters is explicative of about 40% of variance in the data for O_3 and about 12% for CO_2 and Np. Even if particular caution should be used in interpreting these modelling tools in a complex high mountain region like the Karakorum, some robust features have been pointed out. In particular, lower O_3 and higher CO_2 values were observed in concomitance with possible air-mass transport from South Asia and Taklamakan desert, while higher O_3 mixing ratios have been mostly tagged with westerly and northerly air-masses, possibly indicating transport from the free troposphere, while robust evidences for stratospheric intrusions can be obtained only for about 2% of the data-set.

We do not have direct indication concerning the chemical properties, but finding similar particle concentrations at the NCO-P station suggests efficient contamination of the high-altitude regions by emissions located both in the vicinity of the sampling site and further away at regional scale. Our results indirectly support the findings from Qian et al. (2011), who performed a series of experiments using a global climate model and showed that elevated amount of absorbing aerosol (black carbon and dust) can be deposited on the Karakorum snow cover thus favouring significant reduction of the surface albedo, with possible implications on the run-off related to snow melting. As reported by Ming et al. (2008), the transport of absorbing aerosol particles to the upper troposphere and the high altitude glaciers might reduce the net accumulation of snow/ice, further affecting cryosphere and hydrological cycles. We speculate that similar processes can also occur over upper Karakorum, even if further measurements would be necessary to confirm this prospect. Moreover, there is a possibility that, as already observed for the Himalayas (see Bonasoni et al., 2010), the across-valley circulation might represent a “channel” by which polluted air-masses that are richer in O_3 and particles can be effectively transported to the high Karakorum. This would constitute an important issue considering both the direct impact to the mountain ecosystem and biodiversity (see The Royal Society, 2008). Furthermore, the same process can be matter of concern for possible injection of climate forcers to the upper/free troposphere with subsequent climatic impacts at even larger scales (Ramanathan and Carmichael, 2008; UNEP and WMO, 2011).

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.07.063.

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