Tropospheric vertical column densities of NO2 over managed dryland ecosystems (Xinjiang, P. R. China): MAX-DOAS measurements vs. 3-D dispersion model simulations based on laboratory derived NO emission from soil samples

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**Abstract**

We report on MAX-DOAS observations of NO2 over an oasis-ecotone-desert ecosystem in NW-China. There, local ambient NO2 concentrations originate from enhanced biogenic NO emission of intensively managed soils. Our target oasis “Milan” is located at the southern edge of the Taklimakan desert, very remote and well isolated from other potential anthro­po­ge­nic and biogenic NOx sources. Four observation sites for MAX-DOAS measurements were se­lected, at the oasis center, downwind and upwind of the oasis, and in the desert. Biogenic NO emissions in terms of (i) soil moisture and (ii) soil temperature of Milan oasis’ (iii) different land-cover type sub-units (cotton, Jujube trees, cot­ton/Ju­ju­be mixture, desert) were quantified by la­boratory incubation of corresponding soil samples. Net potential NO fluxes were up-scaled to oasis scale by areal distribution and classification of land-cover types derived from satellite images using GIS techniques. A Lagrangian dispersion model (LASAT, Lagrangian Simulation of Aerosol-Transport) was used to calculate the dispersion of soil emitted NO into the atmospheric boundary layer over Milan oasis. Three dimensional NO concentrations (30 m horizontal resolution) have been converted to 3-D (three dimensional) NO2 concentrations, assuming photo­sta­tio­na­ry state conditions. NO2 column densities were simulated by suitable vertical integration of modeled 3-D NO2 concentrations at those downwind and upwind locations, where theMAX-DOAS measurements were performed. Downwind-upwind differences (a direct measure of Milan oasis’ con­tribution to the areal increase of ambient NO2 concentration) of measured and simulated slant (as well as vertical) NO2 column densities show excellent agreement. This agreement is considered as the first successful attempt to prove the validity of the cho­sen approach to up-scale laboratory derived biogenic NO fluxes to ecosystem field con­ditions, i.e. from the spatial scale of a soil sample (cm2) to the size of an entire agricultural ecosystem (km2).

**1 Introduction**

Emissions of nitric oxide (NO) are important in regulating chemical processes of the atmo­sphere (Crutzen, 1987). Once emitted into the atmosphere, NO reacts rapidly with ozone (O3) to ni­tro­gen dioxide (NO2) which, under daylight conditions, is photolyzed back to NO (λ ≤ 420 nm). For that rea­son, NO and NO2 are usually considered as NOx (NOx = NO + NO2). Ambient NOx is a key catalyst in atmospheric chemistry: during the atmospheric oxidation of hydrocarbons its am­bient concentration determines whether ozone (O3) is photochemically generated or de­stroy­ed in the tropo­sphere (Chameides et al., 1992). While the combustion of fos­sil fuels (pow­er plants, vehicles) is still the most important global NOx source (approx. 25 Tg a-1 in terms of mass of N), biogenic NO emissions from soils have been estimated to range between 6.6 and 9.6 Tg a-1 (Denman et al., 2007). The considerable uncertainty about the range of soil bio­gen­ic NO emissions stems from widely differing estimates of the NO emission. Moreover, the un­cer­tain­ties in the NO emission data from semi-arid, arid, and hyper-arid regions are very large (mainly due to a very small number of measurements being available). These ecosystems, however, are considered to contribute more than half to the global soil NO source (Davidson and Kingerlee, 1997), and make approx. 40% of planet Earth's to­tal land surface (Harrison and Pearce, 2000).

Production (and consumption) of NO in the soil depends mainly on soil microbial activity and is mainly controlled by soil temperature, soil moisture, and soil nutrient concentration (Conrad, 1996; Meixner and Yang 2006; Ludwig et al., 2001). Any natural or anthropogenic action that result in the in­puts of nutrients (e.g. by fertilizer application) and/or modification of soil nutrient turnover rates has a substantial effect on soil biogenic NO emission. The rapid (economically driven) int­ensification of arid agriculture (oasis agriculture), par­ti­cu­lar­ly by en­large­ment of the arable area and by enhancement of necessary irrigation leads inevitably to the increase of soil biogenic NO emissions. Since those microbial processes which underlay NO production and NO consumption in soils are confined to the uppermost soil layers (<0.0 5 m depth, Ru­dolph et al., 1996), the most direct method for their characterization and quantification is usually realized by la­bora­tory incubation of soil samples; corresponding measurements result in the determination of so-called net potential NO fluxes, which are explicit functions of soil moisture, soil temperature, and ambient NO concentration (Behrendt et al., 2014).

Tropospheric NO2 column densities can be retrieved from satellite observations using differential optical absorption spectroscopy (DOAS) (e.g. Leue et al., 2001; Richter and Burrows, 2002, Beirle et al., 2004). Identification and quantification of the sources of tropospheric NO2 column densities are important for monitoring air quality, for understanding radiative forcing and its impact on local climate. Ground-based Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) is a novel measurement technique (Hönninger et al., 2004) that represents a significant advantage over the well-established zenith scattered sunlight DOAS instruments, which are mainly sensitive to stratospheric absorbers. From NO2 slant column densities, retrieved from mea­sure­ments at dif­fer­ent elevation angles, information about tropospheric NO2 profiles and/or tro­pospheric vertical column densities can be obtained (e.g. Sinreich et al., 2005; Witt­rock et al., 2004; Wagner et al., 2011).

In this paper we concentrate (a) on ground-based MAX-DOAS measurements of slant and ver­tical NO2 column densities over an intensively used oasis of the Taklimakan desert (NW-Chi­na), (b) on biogenic NO emissions derived from laboratory incubation mea­surements on oasis soil samples, (c) on up-scaling of the laboratory results to the oasis level, (d) calcu­la­ti­on of at­mospheric boundary layer NO2 concentrations by suitable NO→NO2 con­version and 3 dimensional dis­per­sion modelling, and (e) on simulating slant and ver­tical NO2 co­lumn den­si­ties from the calculated 3-D-NO2 distributions by integration along the MAX-DOAS light path. The final aim is comparison and discussion of the results obtained under (a) and (e).

**2 Materials and methods**

**2.1 Research area**

After two ‘searching field campaigns’ (2008 and 2009) in the Xinjiang Uighur Autonomous Region of NW-China, the oasis “Milan” has been identified as the target oasis for the presented re­search. The contemporary oasis Milan, identical to the ancient silk-road post “Miran”, belongs to the county “Ruoqiang” of the Xinjiang province and is located in the southern Taklimakan De­sert on the foot of the Altun Shan Mountains (39.25 °N, 88.92 °E, 998 m a.s.l.). In the early 1950s, the delta-shaped oasis (see Fig. 1) has been established as an agricultural co-ope­ra­tive “state farm” (*Xinjiang Production and Construction Crop*) and covers nowadays about 100 km2. Milan oasis can be geomorphologically classified as a “mountain-oasis-ecotone-desert system (MOED system)” consisting of Gobi (gravel) de­sert, a salty tran­sition zone surrounding the oasis, and dryland farming with irrigation. The latter consists only of two crops, cot­ton and ju­jube trees (*Ziziphus Jujuba* L., “*red date*”), which are planted, irrigated, and fertilized fol­low­ing stan­dar­dized protocols and growing on rectangular fields (approx. 10 ha) of pure cultures or mixtures of it. The general energy supply of Milan oasis is entirely pro­vid­ed by nearby hy­dro­power plants, and battery powered trikes dominate the local public and private transport. Consequently, anthropogenic NOx emissions of Milan oasis are con­sider­ed as very low, if not negligible. Beyond that, Milan oasis is isolated by the desert from neighbouring oases by 80 to 400 km. Therefore, the dominant NOx source of Mi­lan oasis are biogenic NO emissions from its intensively managed crop fields; the oasis can be undoubtedly con­sidered as a large "hotspot in the middle of nothing". Given this very spe­ci­fic situation, it is certainly justified to assume that (a) NO2 concentrations in the atmo­sphe­ric boun­dary layer over Milan oasis are only caused by the oasis itself, and (b) free tropospheric NO2 concentrations, which are usually due to large-scale tro­po­sphe­ric NO2 advection, are negligible.

According to Koeppen classification (Koeppen, 1931; Kottek et al., 2006), Milan oasis owns a cold desert climate (BWk), which is dominated by long hot summers (30 years’ mean: 29°C) and cold winters (30 years’ mean: – 6°C). Mean annual precipitation amounts 28.5 mm, mean annual evaporating capacity is 2920 mm, mean wind direction is NE to E, and mean wind speed is 2.7 m s-1.

***2.2 In-situ* measurements**

A field campaign has been performed at Milan oasis, from 24 May to 26 June, 2011. A total of 32 individual MAX-DOAS measurements (approx. 20 min) have been per­form­ed by two Mini-MAX-DOAS instruments (partially simultaneously) on 21 days during the 2011 cam­paign at the NE natural forest site (1), desert site (2), jujube site (3) and hotel station in Mi­lan oasis center (4). Accompanying data of wind direction, wind speed, air tem­perature, barometric pressure, global and net radiation have been observed at sites (1) – (5) at 1.8 m above ground (at NE natural forest: 11 m; at hotel station: 23 m). Soil temperature (at 0.05 m depth), as well as rainfall (amount and intensity) were recorded at all sites in 2011.

**2.2.1 Ground-based measurements of vertical column densities of NO2**

Multi-Axis-Differential Optical Absorption Spectroscopy (MAX-DOAS) observes scattered sun light under various (mostly slant) elevation angles. From combinations of the retrieved NO2 slant column densities (SCDs) obtained at dif­fer­ent elevation angles, information on the vertical NO2 profile and/or on the corresponding vertical co­lumn density (VCD) can be obtained (e.g. Hönninger et al., 2002; Sinreich et al., 2005; Witt­rock et al., 2004; Wagner et al., 2011). Spectral calibration of the MAX-DOAS instruments was performed by fitting a measured spectrum to a convoluted solar spectrum based on a high resolution solar spectrum (Kurucz et al., 1984). Several trace gas absorption cross sections of NO2 at 294 K (Vandaele et al., 1996), H2O at 290 K (Rothman et al., 2005), Glyoxal at 296 K (Volkamer et al., 2005), O3 at 243 K (Bogumil et al., 2003) and O4 at 286K (Hermans et al., 1999) were convolved to match the resolution of the instrument and then used in the spectral analysis using a wavelength range of 420-450 nm (also a Ring spectrum was included in the fitting process). The output of the spectral analysis is the NO2 SCD, which represents the NO2 concentration integrated along the corresponding light paths through the atmosphere.

Since a spectrum measured in zenith direction (a so called Fraunhofer reference spectrum) is included in the fit process to remove the strong Fraunhofer lines, the retrieved NO2 SCD ac­tually represents the difference between the SCDs of the measurement and the Fraun­hof­er reference spectrum; it is usually referred to as differential SCD or DSCDmeas. The tro­po­sphe­ric DSCD for the elevation angle α can be derived from MAX-DOAS obser­va­ti­on by sub­tract­ing the NO2 DSCD for the closest zenith observation (α0= 90°):

 (1)

DSCDs are converted into VCDs (the vertically integrated concentration) using so called air mass factors (AMF, Solomon et al., 1987), which is defined by:

 (2)

In many cases AMF are determined from radiative transfer simulations (Solomon et al., 1987). However, if trace gas column densities are retrieved from MAX-DOAS observations at high elevation angles (> 10°), the AMF can be determined by the so called geometric appro­xi­mation (Hönninger et al., 2002; Brinksma et al., 2008; Wagner et al., 2010):

 (3)

In this study, the tropospheric vertical column density (VCDtrop) is obtained from DSCDtrop(α) as discussed by Wagner et al. (2010):

 (4)

During the field experiments, the MAX-DOAS instruments have been mounted on solid tables (aluminium struc­ture) at approx. 11 m a.gr. (NW natural forest, hotel station) and 3.5 m a.gr. (remainder of sites) with the telescope facing northwards. Observations were always made on elevation angles of 0°, 2°, 4°, 6°, 8°, 10°, 15°, 20°, 45° and 90°. VCDtrops were determined from measurements at 15°. The potential importance of scattering on the interpretation of the MAX-DOAS measurements depends on two main aspects: first on the height of the trace gas layer and second on the amount of aerosols. In our case the trace gas layer is shallow and the aerosol amount is low (see 2.2.8). Thus scattering effects can be neglected. However, for comparison of the DSCDtrop data obtained by MAX-DOAS with the simulated SCDs obtained from 3 D di­stri­bu­ti­ons of NO2 concentration (calculated with LASAT (Lagrangian Simulation of Aerosol-Transport)) on the basis of laboratory derived net potential NO2 fluxes) the lower elevation angles (2°, 4°) for DSCDtrop(α) have been used, which have a much higher sensitivity to the observed NO2.

For classifying all MAX-DOAS mea­sure­ments whether they were made up-wind, down-wind, or in the center of Milan oasis, their ob­servation po­sition was related to the mean wind direction during each measurement period. Wind measurements were part of accompanying *in-situ* measurements (see below).

**2.2.2 Accompanying measurements**

Wind direction, wind speed, air tem­perature, relative humidity, ba­ro­me­tric pressure, and rainfall intensity have been measured by combined weather sensors (weather transmitter WXT510, Vaisala, Finland). All five weather sensors have been ope­ra­ted side-by-side for one week before they have been mounted at the in­di­vi­du­al measurement sites (1) – (5). Based on these results, all meteorological data, which have been measured between 3 – 24 July, 2011 have been corrected using one of the sensors as reference. All combined weather sensors’ data, as well as those of net radiation (4 component net radiation sensor, model NR01, Hukseflux, The Netherlands) and soil temperature (thermistor probe, model 109, Camp­bell Scientific, U.S.A.) have been recorded every minute. Ambient O3 concentrations and NO2 photolysis rates have also been measured in-situ; both quantities are necessary to calculate the NO→NO2 conversion factor (see Sect. 2.2.8). Ozone concentrations have been mea­sured by UV-absorption spectroscopy (model 49i, Ther­mo­Fisher Scientific, U.S.A.) and NO2 pho­to­ly­sis rate by a filter radiometer (model 2-Pi-JNO2, metcon, Germany) in 1 minute intervals.

**2.2.3 Soil samples**

Microbial processes responsible for biogenic NO emission are confined to the up­per­most soil layers (Galbally and Johansson, 1989; Rudolph et al., 1996; Rudolph and Conrad, 1996). Consequently, composite soil samples (1 kg of top soil, 0–5 cm depth) have been collected at the individual sites of Milan oasis (natural forest, cotton, jujube, cotton & jujube mixture, desert). All samples (air dried) were sent from Xinjiang to Germany by air cargo and stored refrigerated (+ 4°C) until laboratory analysis of the net potential NO flux (see below). Sub-samples have been analyzed for dry bulk soil density (ISO 11272), pH (ISO 10390), electrical conductivity (salinity, ISO 11265), contents of nitrate and ammonium (ISO 14256), total carbon and total nitrogen (ISO 10649 and ISO 13878), texture (ISO 11277), as well of soil water potential (pF values 1.8, 2.5, 4.2, Hartge and Horn, 2009).

Electrical conductivity varied between 1.6 to 9.5 dS m-1 within the managed soils, and was 59.8 and 3.0 dS m-1 in the natural forest and desert soils, respectively. Commercially avail­able soil moisture probes (e.g. TDR (Time-Domain-Reflectometry) and FDR (Frequence-Domain-Reflectometry)) show extreme interferences for soils of >2 dS m-1 (c.f. Kargas et al., 2013) and their calibration for such soils is extremely challenging, if pos­sible at all. Indeed, FDR-signals monitored in Milan oasis’ soils were extremely noisy and spurious. Nevertheless, up-scaling of the laboratory derived net potential NO fluxes needs data of the uppermost layer of each soil of Milan oasis land-types (see Sect. 2.2.6). For that, as most reasonable approximation, it was decided to use that individual (constant) gravi­me­tric soil moisture content, which corresponds to the so-called “wilting point”. The latter was determined by laboratory water tension mea­surements (pF 4.2) on undisturbed soil cores from each land-cover type. The wilting point is de­fined as that soil moisture in the root zone, which would cause irreversible wilting of plants. Wilting point conditions in the uppermost soil layers (2 cm) of soils in the Taklimakan Desert are easily reached, since eva­po­ra­tion is extremely high (eva­po­ra­ting capacity 2920 mm a-1). Even after flooding irrigation of Milan oasis’ crop fields, these conditions have repeatedly been observed within at least 3 days by visual inspections.

**2.2.4 Laboratory determination of net potential NO fluxes**

The methodology for the laboratory measurement of the NO flux from soil has been de­ve­loped at the end of the nineties (Yang and Meixner, 1997) and has been continuously used during the last two de­cades (Otter et al., 1999; Kirk­man et al., 2001; van Dijk and Meix­ner, 2001; Feig et al., 2008a; Feig et al., 2008b; Yu et al., 2008; Ashuri, 2009; Feig, 2009; Gelfand et al., 2009; Yu et al., 2010a, 2010b; Bargsten et al., 2010). The me­tho­do­lo­gy has been significantly improved in the frame of this study and is described in detail by Behrendt et al. (2014).

Generally, the release of gaseous NO from soil is the result of microbial NO production and simultaneous NO consumption. The latter is, as shown by Behrendt et al. (2014), particularly for arid and hyper-arid soils, negligible. Applying the laboratory dynamic chamber method, the release of NO is determined by in­cubating ali­quots of the soil samples in a dynamic chamber sys­tem under varying, but prescribed con­di­ti­ons of soil moisture, soil temperature, and cham­ber’s headspace NO con­centrations. From the difference of measured NO con­cen­tra­tions at the outlets of each soil containing chamber and an empty reference chamber, ac­tu­al net potential NO fluxes (in terms of mass of nitric oxide per area and time) is calculated as func­tion of soil moisture and soil temperature. For that, a known mass (approx. 60 g dry weight) of sieved (2 mm) and wetted (to water holding ca­pa­city) soil is placed in one of six Ple­xiglas chambers (volume 9.7x10-4 m3) in a thermo-con­troll­ed cabinet (0 – 40°C). After pas­s­ing through a puri­fi­ca­ti­on sys­tem (PAG 003, Ecophysics, Switz­er­land), dry pres­surized, zero (i.e., "NO free") air is supplied to each cham­ber, controlled by a mass flow con­­trol­ler (4.167 x 10-5 m3 s-1). The outlet of each cham­ber is connected via a switch­ing valve system to the gas-phase che­mi­lu­mi­nes­cence NO analyzer (model 42i-TL, Thermo Fisher Scien­tific Inc., U.S.A.) and to the non dispersive in­fra­red analyzer CO2/H2O-analyzer (model LI-COR 840A, LI-COR Bio­sciences Inc., U.S.A.). During a period of 24 – 48 h, the soil samples are slow­ly dry­ing out, hence providing the desired variation over the entire range of soil moisture (i.e. from water holding capacity to wilting point conditions and completely dry soil). During the drying out period, the tem­pe­ra­ture of thermo-controlled cabinet is re­peatedly changed from 20 to 30°C, hence providing the desired soil temperature variation (Behrendt et al. 2014). Oc­ca­sio­­nal­ly, nitric oxide standard gas (200 ppm) is di­lu­ted into the air purification system via a mass flow con­troller; this allows the control of the cham­ber headspace NO concentration when determining NO consumption rate of the soil sample. The actual soil moisture con­tent of each soil sample is de­termined by considering the H2O mass balance of each chamber, where the temporal change of the chamber’s head­space H2O con­centration is explicitly re­lat­ed to the evaporation rate of the soil sample. Tracking the cham­ber’s headspace H2O con­cen­tration throughout the drying-out period and relating it to the gravimetrically de­ter­min­ed total soil mass at the start and end of the measurement period delivers the actual gravimetric soil moisture content of the soil sample (Behrendt et al., 2014).

As shown during the last two decades (Yang and Meixner, 1997; Otter et al., 1999; Kirkman et al., 2001; van Djik and Meixner, 2001; van Dijk et al., 2002; Meixner and Yang, 2006; Yu et al., 2008, 2010; Feig et al., 2008; Ashuri, 2009; Feig, 2009; Gelfand et al., 2009 and Bargsten et al., 2010), the de­pen­dence of NO release from gravimetric soil moisture and soil temperature can be characterized by two explicit dimensionless functions, the so-called op­ti­m­um soil moisture curve *g*(θg) and the ex­po­nen­tial soil temperature curve *h*(*T*soil)

 (5)

 (6)

where θg is the dimensionless gravimetric soil moisture content, θg,0 the so-called optimum gra­­vi­me­tric soil moisture content (i.e., where the maximum NO release has been observed), *a* is the soil moist­ure curve’s shape factor (solely derived from NO release and gravimetric soil moisture data which have been observed during the drying-out measurements, see Behrendt et al. 2014), *T*soil is the soil temperature (in °C), *T*soil,0 is the reference temperature (here: 20°C), and *Q*10,NO is the (logarithmic) slope of *h*(*T*soil), defined by

 (7)

where *T*soil,1 is a soil temperature which is 10 K different from *T*soil,0 (here: 30°C). The actual NO fluxes *F*NO (ng m-2s-1; in terms of mass of nitric oxide) are defined by

 (8)

 (9)

where *Q* is the purging rate of the dynamic chambers (m3 s-1), *A*soil is the cross-section of the dynamic chamber (m2), and *m*NO,cham and *m*NO,ref are the NO mixing ratios (ppb) ob­serv­ed un­der conditions (θg,0,*T*soil,0) and (θg,0,*T*soil,1) at the outlets of each soil chamber and the re­fe­ren­ce chamber, re­spec­tively. The conversion of NO mixing ratios to corresponding NO con­cen­tra­tions (ng m-3, in terms of mass of nitric oxide) is considered by *f*C,NO (= 572.5 ng m-3 ppb-1 under STP con­di­tions). Finally, the net potential NO flux, *F*NO(θg,*T*soil) is given by

 (10)

This net potential NO flux is specific for each soil sample, hence for sites (1), (2), (4), and (5) of Milan oasis; the actual NO flux of the sites is calculated applying corresponding field data of gravimetric soil moisture and soil temperature. This procedure has been successfully applied for a variety of terrestrial ecosystems (e.g., Otter et al., 1999; van Dijk et al., 2002; Ganzeveld et al., 2008). For soils of the Zimbabwean Kalahari (Ludwig et al., 2001; Meixner and Yang, 2006), for a German grassland soil (Mayer et al., 2011), but also for Brazilian rainforest soils (van Dijk et al, 2002), soil biogenic NO fluxes derived from the described laboratory incubation method have been successfully verified by field mea­sure­ments using both, field dy­namic cham­ber and micrometeorological (aerodynamic gradient) techniques.

**2.2.5 Classification and actual distribution of Milan fields**

Image classification is likely to assemble groups of identical pixels found in remotely sensed data into classes that match the informational categories of user interest by comparing pixels to one another and to those of known identity. For the purposes of our study, land-cover classification was carried out based on two Quickbird images (0.6 m ground resolution, DigitalGlobe, [http://www.digitalglobe.com](http://www.digitalglobe.com/partners)) acquired on 09 April and 31 August 2007 respectively, with the aid of a recent ETM+ Landsat image (141/033, http://earthexplorer.usgs.gov/) acquired on 25 April 2011 (15 and 30 m spatial resolution). A major advantage of using Quickbird images of high spatial resolution images is that such data greatly reduce the mixed-pixel problem (a “mixed pixel” consists of several land-cover classes) and provide a greater potential to extract much more detailed information on land-cover structures (e.g. field borders, buildings, roads) than medium or coarse spatial resolution data using whether on screen digitizing or image classification.

However, we take the advantage of resolution merge processing to increase the spatial resolution of the Landsat image from 30 to 15 meters for the bands 1-5 and 7 for better land-cover mapping and for updating the land-cover map from 2007 to 2011. Then, we defined different areas of interests (AOIs) to represent the major land-covers with the aid of in-situ GPS data collection (45 points). Next, we increased number of AOIs based on image spectral analysis method. After that supervised classification was performed using the maximum likelihood parametric rule and probabilities. This classifier uses the training data by means of estimating means and variances of the classes, which are used to estimate Bayesian probability and also consider the variability of brightness values in each class. For that, it is the most powerful classification method when accurate training data is provided and one of the most widely used algorithms (Perumal and Bhaskaran, 2010). As a result, five major ecosystems were determined: cotton, jujube, cotton/jujube mixture fields, desert, and plant cover. The cotton and the jujube fields are the most dominant types. Finally, the classified land-cover image was converted into vector format using polygon vector data type to be implemented in LASAT analysis as sources of NO flux and for the purpose of estimating NO concentrations. The map includes 2500 polygons of different sizes as sub-units of Milan major land-cover.

**2.2.6 Two dimensional distribution of soil NO emissions of Milan oasis**

The soil NO emission sources of Milan oasis were defined by individual source units, which have been iden­tified as those sub-units (polygons) of the land-cover vector map consisting of na­tural forest or desert, or covered by cotton, jujube, cotton/jujube mixture. Two identifiers have been attributed to each source unit, (a) a metric coordinate whose numerical format re­fers to the corner of the corresponding polygon, and (b) a unique ID number followed by a de­scription of its land cover type. The soil NO source strength (i.e., actual NO flux, see Sect. 2.2.4) of each source unit has been cal­­culated from the corresponding net potential NO flux, the land-cover type specific gravimetric soil moisture content (“wilting point”), and the actual soil temperature, which has been *in-situ* measured for each of the land-cover types of Milan oasis (see Sect. 2.2.2). Those polygons which are not matching the mentioned land-cover types and other tiny polygons generated by digital image processing techniques were dismissed to avoid intricate geometric errors affecting NO emission data. In other words, these “other classes” were dissolved before performing LASAT analysis to avoid extreme values.

**2.2.7 Three dimensional distribution of NO concentrations by Lagrangian dispersion mo­del­ling (LASAT)**

Having the actual NO source units of the Milan oasis available, the 3-D distribution of NO con­cen­trations in the atmospheric boundary layer (0 – 1500 m a.gr.) over Milan oasis have been cal­culated by the La­gran­gi­an dispersion mo­del LASAT (German VDI Guidelines VDI3945, part 3; c.f. Janicke Consulting, 2011). LASAT is a state-of-the-art model, since (a) LASAT is one of those transport-dispersion models of air-pollution which is officially licensed for legal use of environmental issues (in Germany), and (b) among comparable micro-scale (e.g. street canyons) transport-dispersion models LASAT considers at least chemical transformations of 1st order and keeps nonetheless truly operational. Being a transport-dispersion-model, LASAT basically considers advection (“pixel cross-talk”) applying the 3D-continuity equation for any chosen tracer (see German VDI Guidelines VDI3945, part 3, cf. Janicke Consulting, 2011). For that, pre-processing of meteorolo­gi­cal pa­ra­meters (i.e. 3-D wind distributions, based on meteorological in-situ mea­sure­ments, see Sect. 2.2.2) and calculation of dispersion parameters (σy, σz) have to be performed. Unfortunately, it was difficult to obtain fine resolution using LASAT individually. Therefore, LASAT model was integrated with Geographic Information System (ArcGIS) by using an advanced mo­dule namely LASarc (IVU Umwelt GmbH, 2012). LASarc allowed us to calculate NO concentrations using relatively fine resolution of 30m×30m and taking the advantages of using integrated map colour scheme in ArcGIS. This module has been used to realize Milan oasis’ complex NO source configuration and to setup cal­cu­la­tions of LASAT.

The model was designed to calculate NO-concentrations at 16 different vertical layers (0–3, 3–5, 5–10, 10–20, 20–30, 30–50, 50–70, 70–100, 100–150, 150–200, 200–300, 300–400, 400–500, 500–700, 700–1000, and 1000–1500 m a.gr.). The horizontal resolution is 30 m, in x-direction (W-E) as well in y-direction (S-N), which results in 656 (x) and 381 (y) grids for the Milan oasis domain. LASAT’s meteorological input data contain a variety of pa­ra­meters, namely start and end time (T1, T2), wind speed (Ua) and wind direction (Ra) at ane­mo­me­ter height (Ha), average surface rough­ness (Z0), and atmospheric stability (in terms of stability classes). These pa­rameters have been provided in a time-dependent tabular form, up-dated every 30 minutes (except Z0). Average (30 min) wind speed and wind direction data have been calculated from *in-situ* measurements (1 min resolution, see Sect. 2.2.2).

LASAT’s pre-processing module determines the vertical profile of wind speed according to the well-known logarithmic relation,

 (11)

where *U(z)* is the horizontal wind speed (m s-1) at height *z* (m), *u\**is the friction velocity

(m s-1), *k* is the dimensionless von Karman constant (= 0.4, Simiu and Scanlan, 1996), and *Z0*is the surface roughness length (m). LASAT’s pre-processing module accepts only one in­di­vi­du­al value for *Z*0; nevertheless, the required mean value has been calculated from all *Z*0’s of Milan oasis domain, which have been assigned to each of the sub-units (polygons) of the vector land-cover map (see Sect. 2.2.5). For individual *Z*0’s, we calculated land-cover specific NDVI data (normalized differential vegetation index) from Landsat ETM+ image (141/033)

 (12)

where *NIR* is the reflectance in the near-infrared bandwidth (0.77-0.90 µm) and *RED* is the reflectance in the red bandwidth (0.63-0.69 µm). In Landsat ETM+ images, these corre­spond to bands 4 and 3, respectively. Finally, *r*NIR and *r*RED are the corresponding ratios of reflected and incident energy as a function of wavelength (see Chander and Markham, 2003). Then, surface roughness grid data was estimated as:

 (13)

where *a*xy and *b*xy are constants, which are, according to Morse et al. (2000), derived from *NDVI*(*x,y*) and GPS(x,y) data for known sample pixels representing the earlier classified land-cover types, namely natural forest, desert, cotton, ju­jube, and cotton/jujube mixture. Corresponding land-cover type *Z*0’s are 0.45, 0.01, 0.18, 0.26, and 0.22 m, respectively; the required average value over the entire LASAT model domain results in *Z*0 = 0.22 ± 0.158 m.

Besides mechanical turbulence (*Z*0), atmospheric stability affects most the dispersion of trace substances. For Milan oasis’ atmospheric boundary layer, atmospheric stability has been cal­cu­lated according to the “solar radiation/delta T (SRDT)” method in 30 min intervals. This me­thod (c.f. Tur­ner, 1994) is widely accepted because of its simplicity and its represent­a­tive­ness for atmosphe­ric stability over open country and rural areas, like the Milan oasis domain. Daytime stability classes are calculated from *in-situ* measurements of solar radiation and horizontal wind speed (see Sect. 2.2.2).

Finally, 30 min means of all parameters and input variables of LASAT have been calculated. Using these, about 4×106 gridded data points of 3-D NO concentration have been calculated for each time period considered in Section 3.2.

**2.2.8 Simulation of SCD**NO2 **and VCD**NO2 **by spatial integration of LASAT results**

There is only one tool to provide a robust relationship between biogenic soil NO emissions on one hand and MAX-DOAS observed SCD‘s and VCD’s on the other hand: the exact si­mu­la­tion of the MAX-DOAS measurement through spatial integration of three dimensional NO con­­cen­trations calculated by LASAT (followed by NO→NO2 conversion). At a given location of the MAX-DOAS measurement, in­te­gra­tion must be per­for­med from the height where the MAX-DOAS in­stru­ment has been set-up (*h*MAXDOAS) to the end of the at­mo­spheric boundary layer (*h*ABL = 1500 m a.gr.) along two virtual light paths, (a) the vertical up path (VCD), and (b) the slant path (SCD) according to the selected elevation angle of each MAX-DOAS measurement.

Calculation of simulated VCD’s for NO (VCDNO,sim) at the location of a MAX-DOAS instrument is achieved as follows: (a) determination of the NO mass density (ng m-2) of the ver­tical co­lumn between *h*MAXDOAS and *h*ABL; this is obtained by adding NO concentrations (ng m-3 in terms of mass of nitric oxide) of all LASAT cells in vertical direction over that

30 m × 30 m grid, which contains the location of the MAX-DOAS instrument, multiplied by the height difference Δ*h* = *h*ABL− *h*MAXDOAS (in m), (b) mul­ti­plying that NO mass density by the ratio of Avogadro’s num­ber (6.02217×1026 molecules kmol-1) and the molecular weight of NO (30.0061×1012 ng kmol-1) delivers the desired value of VCDNO,sim in units of molecules

m-2 (×10-4: mo­le­cul­es cm-2) at the location of the MAX-DOAS instrument. Calculation of simulated SCD’s for NO (SCDNO,sim) requires the de­ter­mi­na­ti­on of the 3-D light path through the trace gas layered. Positioning of MAX-DOAS’s te­le­scope was always to the north, the selected MAX-DOAS elevation angle α and *h*ABl deliver the length of the slant light path (= *h*ABL/sinα). The de­sired SCDNO,sim (in molecules m-2) re­sults from the NO mass density of the slant column multiplied by the length of the slant light path, where the NO mass density is equivalent to the sum of all NO concentrations of those LASAT cells which are intersected by the slant light path from the position of the MAX-DOAS instrument to *h*ABL.

For conversion of VCDNO,sim to VCDNO2,sim and SCDNO,sim to SCDNO2,sim it is assumed, that the pho­tostationary state (PSS) of the triad NO, NO2, and O3 is established in Milan oasis’ atmo­sphe­ric boundary layer. According to Leighton (1961) this chemical equilibrium state is due to fast photochemical reactions, namely NO+O3→NO2+O2 and NO2+hυ→NO+O, from which the so-called pho­tostationary state NO2 concentration (NO2,PSS) can be derived as

 (14)

where [O3] is the ozone number density (molecules cm-3; calculated from *in-situ* measured O3concentrations, see Sect. 2.2.2), [NO] is the NO number density, *k*1 is the reaction coefficient of the NO+O3→NO2+O2 reaction (cm3 molecules-1 s-1; Atkinson et al., 2004), and *j*(NO2) is the *in-situ* measured NO2 photolysis rate (in s-1; see Sect. 2.2.2). Finally, VCDNO2,sim and SCDNO2,sim are cal­cu­la­ted from VCDNO,sim and SCDNO,sim by

 and  (15)

where the NO→NO2 conversion factor is defined by *CF*0 = [O3] *k*1 / *j*(NO2).

Since the NO-NO2-O3 photochemical equilibrium could not be handled by LASAT’s “chemical” algorithm, we decided to use measured data (O3 mixing ratio, NO2 photolysis rate, s. sect. 2.2.2) to convert the calculated 3D-NO mixing ratio to the photo-stationary 3D-NO2 mixing ratio. For that, a constant vertical O3 mixing ratio (up to 1500 m a.gr.) is assumed over Milan oasis. This is justified by the fact, that particularly in arid and hyper-arid landscapes at mid-day conditions (maximum of insulation) the entire atmospheric boundary layer is intensively mixed, which is due to extensive convective heating of the surface by the sun which produces powerful buoyant thermals that establish the so-called mixing layer. Consequently an uniform vertical mixing ratio is expected for trace gases with chemical lifetimes greater than the exchange time of the atmospheric boundary layer. (c.f. Husar et al. 1978, Stull 1988). This assumption is valid for ozone. Vertically constant O3 mixing ratio has been reported for the atmospheric boundary layer over semi-arid southern Africa (Meixner et al., 1993). Concerning the vertical distribution of j(NO2) it is obvious, that the downward component of the actinic flux increases with increasing elevation due to the decreasing optical thickness of the scattering air masses. However, the altitude effect on the actinic flux in the first kilometer of the troposphere is typically very small. Trebs et al. (2009) used the Tropospheric Ultraviolet Visible model to calculate the typical vertical change of the actinic flux and found a vertical gradient of 1.1%/km. Consequently, our calculations of the NO to NO2 conversion in the boundary-layer over Milan oasis (1500 m a.gr.) have not considered any potential vertical change of the j(NO2) values measured at ground level. Nevertheless, for the case of our measurements the locally enhanced NO values caused by the soil emissions have a small but systematic effect on the ozone concentration, and thus also on the Leighton ratio: Close to the surface (below about 50m) the NO concentrations can be quite large, with maximum values up to about 10 ppb. Consequently, the ozone concentration will be reduced due to the reaction with NO by up to about 10 ppb. This means that the Leighton ratio will be reduced by up to about 25%. Although the reduction of the ozone mixing ratio wil be partly compensated by mixing with air from higher altitudes, the simulated NO2 mixing ratios might overestimate the true NO2 mixing ratios by up to about 25%. Probably the true overestimation for our measurements is much smaller because the typical NO mixing ratio within the lowest 100m is much lower than 10 ppb.

**3 Results and Discussion**

**3.1 Land-cover type specific net potential NO fluxes**

Net potential NO fluxes (as functions of soil temperature and moisture) have been de­ter­min­ed by incubation of samples which have been taken from the top-soil of Milan oasis’ major land-cover types, i.e. natural forest, desert, cotton, ju­jube, and cotton/jujube mixture (see Sect. 2.2.4). Figure 2 shows the laboratory derived net potential NO flux, *F*NO from soils of the most contrasting land-cover types of Milan oasis (irrigated & fertilized fields of cotton, jujube, cotton/jujube mixture, and desert).

Net potential NO fluxes of the natural forest land-cover type are not shown, because laboratory incubation measurements have shown that there is no significant NO release from these soils, most likely due to its high electrical conductivity (salt content). Optimum gravimetric soil water contents (i.e., where the maximum of *F*NO is observed) for desert, managed cotton, and managed jujube soils have one in common, very low values of θg,opt (0.009−0.017) for soil temperatures of 50°C. During the vegetation period (April − September), soil temperatures of > 40°C are easily reached for the soils of Milan oasis, particularly for the desert soils. While the nature of all Milan oasis’ soils is arid/hyper-arid, maximum net potential NO fluxes are 7600, 63, 270, and 98 ng m-2 s-1 (in terms of mass of nitric oxide) for cotton, jujube, jujube/cotton mixture, and desert soils, respectively.

**3.2 Land-cover types of Milan oasis and actual NO fluxes**

As mentioned in Section 2.2.5, land-cover classification and actual distribution of Milan oasis’ fields have been identified from satellite images (Quickbird, Landsat ETM+). The 2011 distribution of fields and the corresponding land-cover is shown in Figure 3.

The dominant crop was cotton, representing 18% (64 km2) of the total field area of Milan oasis (jujube 7%, 28 km2), cotton/jujube mixture 0.89 % (3 km2), natural forest 18% (64 km²), residential area 1.62% (5.5 km²) and desert 52% (174 km2). Land-cover specific, actual NO fluxes (30 min means) from cotton, jujube, cotton/jujube, and desert soils were calculated from corresponding laboratory derived net potential NO fluxes, land-type specific soil moisture and soil temperature data (see Sect. 2.2.6). These NO fluxes (ng m-2s-1, in terms of mass of nitric oxide) were then assigned to each individual source unit (i.e. to each of the 2500 polygons of Milan oasis’ domain).

For the period 03 to 24 June, 2011, land-cover specific, actual NO fluxes were calculated

according to eq.(10) for cotton, jujube, cotton/jujube, and desert soils from corresponding laboratory derived net potential NO fluxes. As input we used land-type specific, measured soil temperature data as well as land-type specific soil moisture data (so-called “wilting points”, s. Sect. 2.2.3). The calculated NO fluxes are shown in Fig. 4 as median diel variation (for the entire period of 03 to 24 June, 2011). Since NO fluxes from Milan cotton fields dominate the total soil biogenic NO emission of the oasis, corresponding medians and quartiles are shown in Fig. 4, while – for the sake of clarity – for jujube, cotton/jujube, and desert only medians are given. Since land-type specific “wilting points” are constant, diel variations of actual NO fluxes mirror directly those of corresponding soil temperatures, showing the daily minimum around 06:00 local time for all four major land-cover types. The maximum of the actual NO-flux, however, is around 13:00 (local time) for jujube, cotton/jujube, and desert soils, and 15:00 local time for cotton. This is due to the growth of the cotton plants: while at the beginning of the experimental period the bare soil surface was nearly 100% exposed to insolation, the growing cotton canopy has shaded great parts of the soil surfacetowards the end of the experimental period . This is also reflected by the skewed distribution of actual NO-fluxes from cotton covered soil, indicated by the daytime non-symmetric inter-quartile range (= upper quartile   lower quartile). As shown in sect. 3.5, actual NO-flux data of 09 June, 2011 (08:30-14:30 local time) were used for the comparison of LASAT and MAX-DOAS results. During this particular day (within the first week of the experimental period), the derived flux for “land-cover cotton” ranged from 15–64 ng m−2s−1 (in terms of mass of NO), those for jujube, cotton/jujube, and desert land-covers ranged from 11–13, 6–16, and 6–17 ng m−2s−1, respectively. These actual NO fluxes were then assigned to each individual source unit (i.e. to each of the 2500 polygons of Milan oasis’ domain). The soil biogenic NO emission from all cotton fields between 08:30 and 14:30 was estimated to 28.7 kg (in terms of mass of NO), equivalent to 76% of the total soil biogenic NO emission of the entire Milan oasis within 6 hours.

**3.3 Vertical NO2 column densities by MAX-DOAS**

We performed 32 individual MAX-DOAS measurements within 21 days of the 2011 field campaign to examine the spatial variation between the observed sites. In Fig. 5, all observed ver­tical NO2 column densities (in molecules cm-2) observed at sites (1) - (4) of Milan oasis are shown in polar coordinates with reference to corresponding wind directions measured *in-situ* at the individual sites.

Wind speeds (30 min means) ranged between 1.5 and 7.7 m s-1 and wind direction was mostly (78%) from the northern quadrants (59%, 9%, 13%, and 19% from NE, SE, SW, and NE quadrants, respectively). As expected, highest VCDs (1015 –1016 molecules cm-2) were observed at site (4) (Milan oasis center), regardless of wind direction. When the wind direction is from the NE quadrant, site (3) (ju­jube fields) is down-wind of Milan oasis (see Fig.1); then its VCDs are as high as those ob­tain­ed in the oasis’ cen­ter (5−7×1015 molecules cm-2). The few VCD data points of 1×1015 molecules cm-2 at the jujube site, attributed to winds from SE and SW quadrants, are mainly due to NO emis­si­ons from traffic on the National Road 315 which passes the southern margins of Milan oasis. Lowest VCDs (3×1013−3×1014 molecules cm-2) have been observed at site (1) (natural forest) and site (2) (desert). Alone from these spatially resolved VCD observations in the Milan oasis’ domain, the increase of VCD due to the oasis itself can be estimated in the order of at least one order of magnitude.

Fortunately, we have been able to perform simultaneous measurements with two MAX-DOAS instruments at sites (1) and (3) on 09 and 13 June, 2011. Since winds (approx. 3 m s-1) were from the NE quadrant during these two days, site (1) has been up-wind, and site (3) downwind of Milan oasis. Corresponding VCD results are shown in Figure 6. NO2 VCDs at the downwind site exceeded those at the upwind site by factors 5 – 9. This dif­fer­ence between downwind and upwind MAXDOAS signals is considered to be a direct measure for the areal increase of am­bient NO2 concentration. In the absence of an­thro­po­ge­nic NOx sources (see Sect. 2.1), this provides first evidence for the considerable impact of the bio­genic NO emis­si­ons from the fields of Milan oasis.

**3.4 3-D distribution of ambient NO-concentration**

The LASAT model has to be used to calculate the dispersion of soil emitted NO into the at­mo­spheric boundary layer over Milan oasis. An example for the resulting di­stribution of NO con­­centration in the first four vertical layers of LASAT (0-3, 3-5, 5-10, and 10-20 m) is shown in Figure 7 (09 June, 2011; 11:30-13:00 local time). The shown results are the mean of three LASAT model runs, since a new LASAT calculation of 3-D di­stribu­ti­on of NO concentration is star­ted for every set of meteorological parameters which are provided every 30 min from means of the *in-situ* measured meteorological quantities (see Sect. 2.2.2). During 11:30-13:00, mean wind direction was 15°, 38°, and 50°, wind speed was rather constant (2.60 – 2.67 m s-1), and atmospheric stability class has been generally neutral (3.2).

By comparing the NO ambient concentrations, particularly in the first vertical LASAT layer (0−3 m) of oasis area with the surrounding desert, it becomes obvious that the great dif­fer­enc­es of ambient NO concentrations mirror the corres­pond­ing differences of actual soil NO fluxes from each source-unit; within this layer calculated mean NO concentrations are 13, 12, 10, and 1 ng m-3 (in terms of mass of nitric oxide; or 10.6, 9.8, 8.2, and 0.8 ppb) for the oasis centre, jujube fields, cotton/jujube mixture, and desert, respectively. The value at the oasis center exceeds those over desert by more than an order of magnitude, similar as the corresponding VCD values (see above). As expected under the prevailing conditions of well developed atmospheric turbulence, NO concentrations rapidly decreases with height (see panels “0−3 m”, “3−5 m”, “5−10 m” in Fig. 7), and with prevailing northerly winds, the NO con­cen­­tration centre shifting southwards with increasing altitude.

**3.5 Simulated SCDs and VCDs *vs.* SCDs and VCDs by MAX-DOAS**

For those periods where simultaneous “upwind” and “downwind” MAX-DOAS measurements have been performed (09 and 13 June, 2011), corresponding SCDsim and VCDsim have been simulated by suitable vertical integration (see Sect. 2.2.8) of LASAT-calculated 3-D NO con­cen­tra­ti­ons, fol­low­ed by NO→NO2 conversion (based on photostationary state assumption of Milan oasis’ atmospheric boundary layer). Since SCDsim and VCDsim represent only that part of true SCDs and VCDs of NO2, which are due to the contribution of the oasis’ soil NO emissions, SCDsim and VCDsim are compared to the difference of those SCDs and VCDs which have been simultaneously measured by two MAX-DOAS instruments at corresponding “downwind” and “upwind” sites (see Fig. 8). For elevation angles of 2° and 4°, SCDsim and ΔSCD = SCDdown − SCDup are shown in Figure 8a. In Figure 8b, VCDsim and ΔVCD = VCDdown − VCDup are shown for 15° elevation.

Here it should be noted that in principle the accuracy of the geometric approximation is higher for the high elevation angles than for the lower elevation angles. However, for the specific cases studied here, this is not the case. First, close to the sources, the height of the layer with elevated NO2 is quite low (in our case the bulk of NO2 is located below 100 m). Second, also the aerosol load is usually very low. Thus the probability of scattering events inside the layer of enhanced NO2 is very low, and consequently the accuracy of the geometric approximation is relatively high. To further quantify the associated uncertainties, we performed radiative transfer simulations and found that the deviations from the geometric approximation are similar for the different elevation angles (about 5% for 2°, 3% for 4° and 3% for 15°). However, because of the shorter light paths through the NO2 layer, the relative error caused by the uncertainty of the spectral analysis is higher than for the low elevation angles. Thus for the case of our measurements, we indeed expect lower uncertainties for the low elevation angles.

Since soil NO emission data used in the LASAT dispersion model were calculated from land-cover type specific potential net NO fluxes, which in turn were derived from la­bo­ra­tory incubation experiments on corresponding soil samples, the results in Figure 8 are also considered as an excellent quality assurance of the chosen up-scaling of laboratory results to the oasis scale. There is remarkable good agreement between measured and si­mu­la­ted data.

However, the actual NO emissions (irrespective of the land-cover type) have their maximum in the early afternoon (s. Fig. 4), while the highest height-integrated NO2 concentrations as simulated by LASAT (on the basis of the actual NO emissions) are in the morning (08:30–10:00), followed by rather constant values for the reminder of the day (s. Fig. 8). The apparent discrepancy between both diurnal variations can be simply explained by the diurnal variation of the wind direction and the specific viewing geometry of the MAX-DOAS instrument. The MAX-DOAS instrument was located at the south-west corner of the oasis, and the observations at zenith and low elevation angles probed air masses located at different locations accross the oasis. The wind direction was from north-east in the morning and turned to north-west in the afternoon. Hence, air masses of lower concentration crossed the viewing directions in the afternoon compared to those in the morning. This explains that in spite of the larger NOx emissions smaller column densities have been observed in the afternoon. The apparent discrepancy of the diurnal cycles of NO emissions and measured NO2 column densities indicates the importance to exactly consider the 3-dimensional NO2 distribution (due to the soil-emitted NO) for the comparison of the model results with MAX-DOAS observations.

The Figure 8b shows that the LASAT simulations overestimate slightly the true NO2 VCD. The both measured and simulated NO2 VSDs have with an average root mean square (RMS) error between the measured and simulated values of approx. 5-15%. However, the overestimation of LASAT simulation is well suited to the fact that in reality a little less NO can be converted to the NO2 because of lower ozone concentration at the surface. **4 Conclusion**

This study has been focused on the following activities: (1) representative soil sampling from the up­per­most soil layer (< 0.05 m) of all land-cover type units (natural forest, cotton fields, jujube fields, cotton/jujube mixture, desert) of Milan oasis (Xinjiang, NW China), (2) la­bo­ra­to­ry in­cu­ba­tion experiments (dynamic chamber system) to characterize the bio­genic NO emis­si­on from these soil samples in form of net po­ten­tial NO fluxes as function of soil mois­ture and soil temperature, (3) determination of the actual size, areal distribution, and land-cover type of Mi­lan oasis’ field units from satellite remote sensing information, (4) field mea­sure­ments of slant (SCD) and vertical (VCD) NO2 column den­sities (by MAX-DOAS) and additional quantities (soil moisture, soil temperature, ozone con­centration, NO2 photolysis rate, meteorological parameters) during an extended field cam­paign of 4 weeks at Milan oasis, (5) using data from (2), (3) and (4): calculation of Milan oasis’ 2D distribution of actual, land-cover specific NO fluxes, (6) calculation of 3-D NO con­cen­trations in Milan oasis’ atmo­sphe­ric boundary layer originating from the dispersion of biogenic NO soil emis­sions determined by (5) with help of the Lagrangian dispersion model LASAT, (7) si­mu­la­tion of SCDs and VCDs by suitable vertical integration of calculated 3-D NO con­cen­tra­tions fol­lowed by suitable NO→NO2 conversion factors derived from *in-situ* measurements, (8) com­parison of measured and simulated SCDs and VCDs.

Results of the laboratory derived NO fluxes have shown that the extensively managed (fer­ti­li­zed and efficiently irrigated) cotton fields of Milan oasis release large amounts of soil bio­genic NO; NO fluxes range between 10−30 ng m-2 s-1 (in terms of mass of N), that is approx. 5−10 times more than from a typical central European wheat field (Yamulki et al. 1995; Stohl et al. 1996).

Applying two MAX-DOAS instruments, simultaneous measurements have been performed at upwind and downwind sites of Milan oasis. Downwind site VCDs exceeded those from the upwind site by factors 5 – 9. Differences of VCD and SSC (“downwind” minus “upwind”) are a di­rect measure for the areal increase of ambient NO2 concentration caused by the oasis itself. The measured differences of VCDs and SCDs were compared with the si­mu­lated VCDs and SCDs and excellent agreement was found.

This agreement is considered as the first successful attempt to prove the validity of the cho­sen approach to up-scale laboratory derived biogenic NO fluxes to ecosystem level field con­ditions, i.e. from the spatial scale of a soil sample (cm2) to field size (ha), and from field size (ha) to the size of an entire (agro-) ecosystem (km2). Furthermore, in the absence of an­thro­po­genic NO sources of Milan oasis (hydropower energy, battery powered trikes), it is obvious, that the areal increase of ambient NO2 concentration in the atmospheric boun­dary layer of the isolated (in terms of NO2 advection) Milan oasis is entirely due to biogenic NO emission from the arid/hyper-arid soils of the oasis itself. Extensive agricultural manage­ment of Milan oasis’ crop fields (fertilization (350˗600 kg N ha-1a-1) and effective irrigation of cotton and jujube fields) obviously provides considerable contribution of biogenic NOx (NO+NO2) from arid/hyper-arid soils of the Taklimakan desert to the local tropospheric NOx budget.

About 80% of the Chinese cotton production originates from the 3000 km long belt of oases sur­rounding Taklimakan Desert (1.65×106 km2) in Xinjiang (NW-China); cotton cultivated land area in Xin­j­i­ang occupies the first place of entire China. Since 1955, Xinjiang's output of cot­ton in­creas­ed 294 times (Lei et al., 2005). Fast economic growth in the region (+11% GDP a-1), inevitably ac­com­pa­nied by large anthropogenic NOx emissions (traffic, energy pro­duc­ti­on), may be coun­ter­vail­ed or even exceeded by the “hotspot” character of Xinjiang’s oases, namely by soil biogenic NO emissions from agriculturally dominated oases. Most likely, they will contribute most to the re­gional tropospheric NOx budget. This is all the more likely, given the continued in­ten­si­fi­ca­tion of oasis agriculture around the Taklimakan desert which will be accompanied by corresponding land use change (desert→dryland farming with irrigation) in the coming decades.

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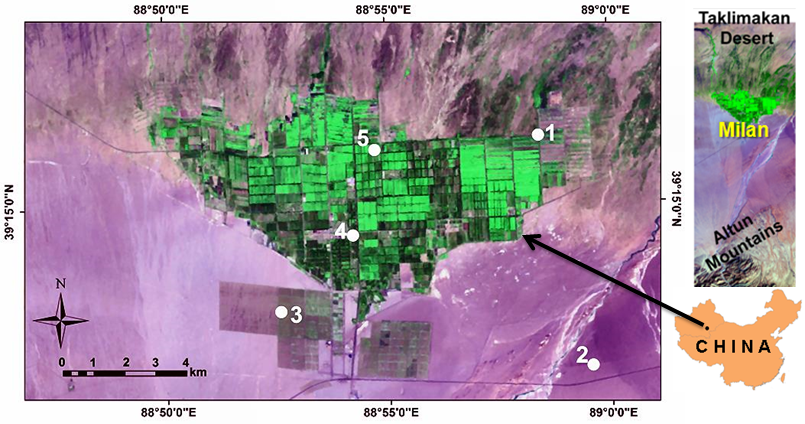
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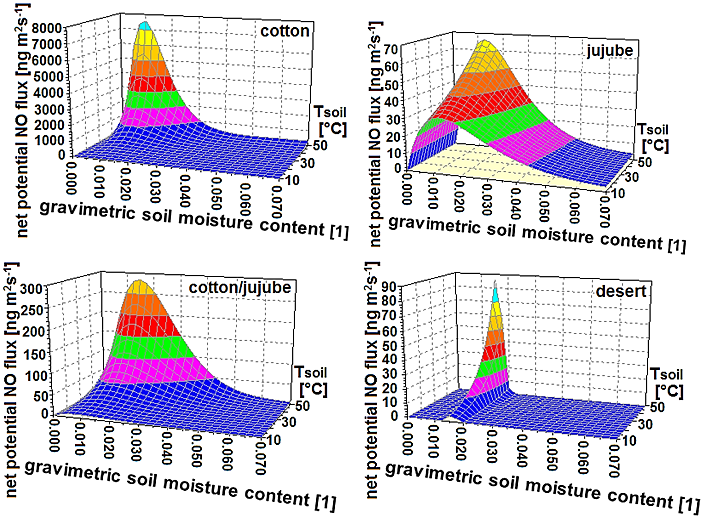
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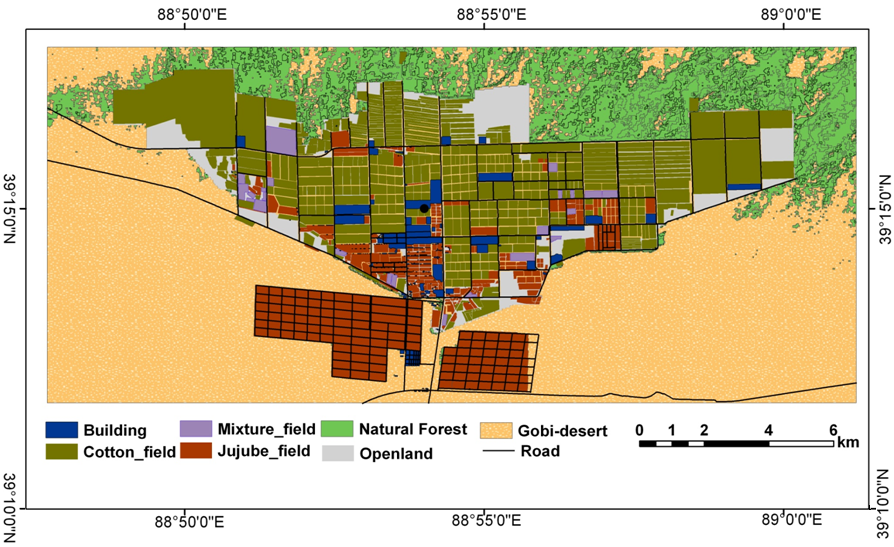
**Figures:**



**Fig. 1**: Satellite map (Landsat ETM+; 2011) of Milan oasis, Xinjiang, NW-China (The map has an area of 338 km2). The white circles show the sites of *in-situ* measurements: natural forest (1), desert (2), jujube (3), hotel/oasis station (4) and cotton field (5).



**Fig. 2**: Net potential NO fluxes *F*NO (ng m-2 s-1; in terms of mass of nitric oxide) from soils of the four major land-cover types of Milan oasis as functions of soil temperature (°C) and dimensionless gravimetric soil moisture content.



**Fig. 3**: 2011 map of land-cover types of Milan oasis as derived from satellite images (Quickbird, Landsat ETM+, see Sect. 2.2.5).

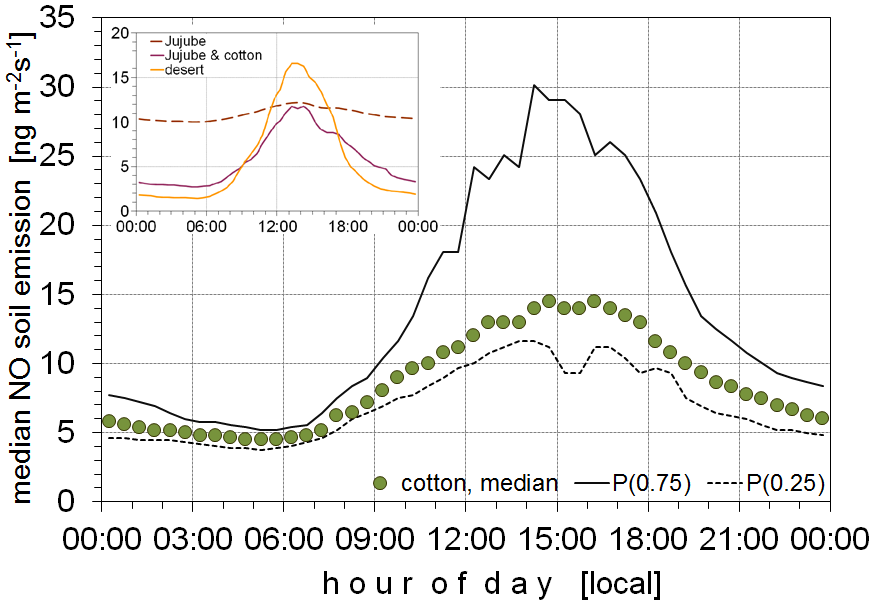
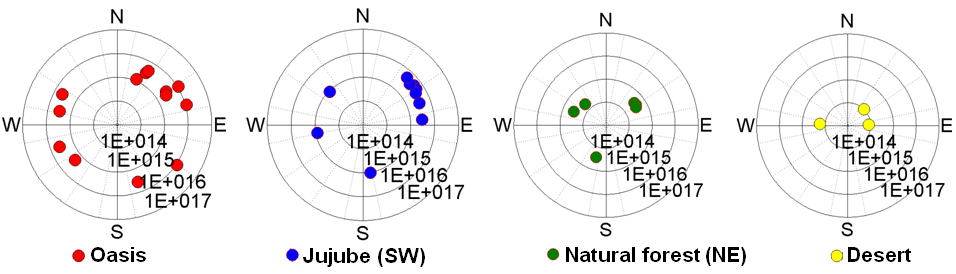
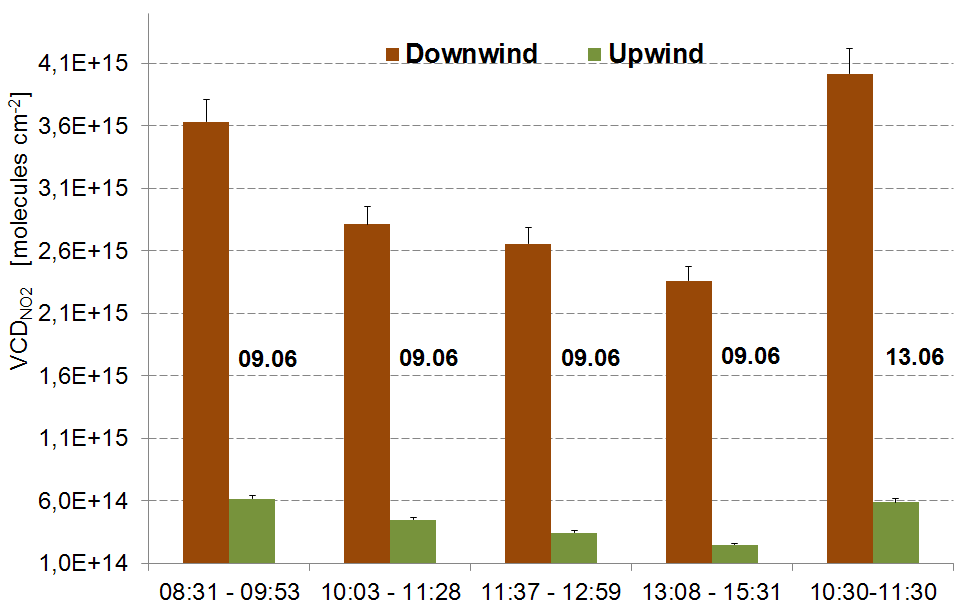
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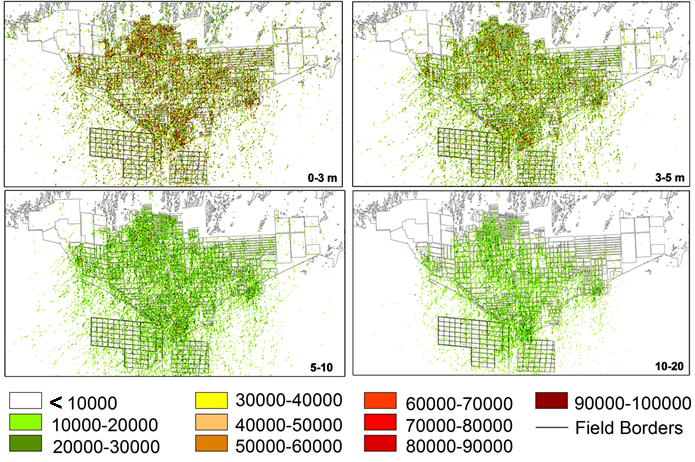
Fig 4:median diel variation of the actual NO-flux (ng m−2s−1; in terms of mass of nitric oxide) from soils of the four major land-cover types of Milan oasis for the period 03 to 24 June, 2011. Data have been calculated according eq.(10) using (a) soil temperatures (medians) measured for each of the four major land-cover types, and (b) so-called “wilting point”-data for corresponding soil moisture contents at the four sites (s. section 2.2.3). Data for the cotton-site are given as medians, as well as 25 and 75% quantiles, those for the Jujube, Jujube-cotton and desert sites as medians only (s. figure insert).



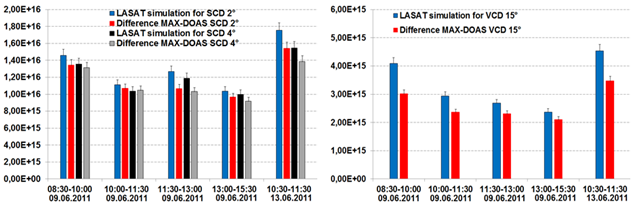
**Fig. 5**: Results of MAX-DOAS measurements performed at sites oasis/hotel (4), Jujube (3), Natural forest (1), and Desert (2) of Milan oasis from 23 May to 26 June, 2011 (see Fig. 1). Ver­tical NO2 column densities (in mole­cules cm-2; 20-30 min averages) are shown in relation to in-situ measured wind di­rec­ti­on at each location of MAX-DOAS mea­sure­ments. The MAX-DOAS mea­sure­ments were performed between 6:00 and 19:00 (local time). Note the radial lo­ga­rith­mic scale of VCD data.

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**Fig. 6**: Results of NO2-VCD measured simultaneously with two MAX-DOAS instruments up­wind (natural forest, site (1)) and downwind (jujube field, site (3)) of Milan oasis on 09 and 13 June, 2011.



**Fig. 7**: Results of NO concentrations (ng m-3; in terms of mass of nitric oxide) calculated by the LASAT dispersion model for the first four vertical levels on 09 June, 2011, 11:30 to13:00 (local time).

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**Fig. 8**: Simulated SCDs vs. SCDs measured by MAX-DOAS (a) and simulated VCDs vs. VCDs measured by MAX-DOAS (b) on 09 and 13 June, 2011 at Milan oasis. SCDs have been measured and simulated for elevation angles of 2° and 4°, VCDs were measured at 15°.