We sincerely thank both reviewers for providing her/his very valuable suggestions, which helped us improve our paper significantly. Below please find our responses:

Reply to Anonymous Referee #1

General comments:
(1) Mobile measurements: More details would be very helpful. It's not clear at all how many, when, and what routes were taken for the mobile measurements. The authors simply state that mobile measurements "were performed in June and July 2016" and that "varying routes" were taken. Were mobile deployments performed every day over those two months? What were the routes like? I suggest the authors at least provide a table of the dates and times that mobile measurements were made, and a map of each of the routes (unless they were all identical, or the number of mobile deployments was truly large) as Supplementary Information, and include a short description in the main text (at least N=?). Since I assume the route was not identical in each case, might be useful to include a panel in Figure 3 showing how many spatially overlapping measurements are included in each pixel average (i.e. how many days are included in each 20 x 20m average?). Also, it's not clear until much later in the manuscript that the authors explain these observations are gridded to 20 x 20 m. This should be included in the description here.
As the reviewer suggested, detailed description regarding hours per day of all the measurement days and the total amount of measurements per day have been listed in Table 1, Section 2.1.

All measurements were spatially averaged to a high-resolution grid of 20m x 20m. This resolution has been chosen according to an average driving speed of 40-50 km/h and the measurement sampling time of 2s, so that at least one measurement value per drive-by falls into each grid box. All measurements from different drives and different days are averaged for each grid cell after diurnal cycle normalization.

This description was added in the paper (Section 2.1, line 10-15).

(2) Long path DOAS: I'm also a little confused about how all the different LP-DOAS measurements were used. The methods describe four different absorption paths, shown in Figure 1. But when/how is each absorbing path used? It seems to me that the LP-DOAS measurements are only used in the normalization step (Section 3.1.1).3 Were all the LP-DOAS measurements averaged together to get each day's diurnal cycle? If so, how? Or were different absorption paths used for different parts of the city? Or were the results from only one absorption path used? (If the latter, why are all paths described and shown?)
For the long-term average diurnal cycle calculation described in Sec.3.1.1 all available data from all measurement paths were used. LP DOAS measurements of light paths towards to Hilton hotel, TUM University, and LMU physic department were used to normalize the mobile measurements regarding the diurnal cycle. The same description was added in the paper (Section 2.2, line 19-22).

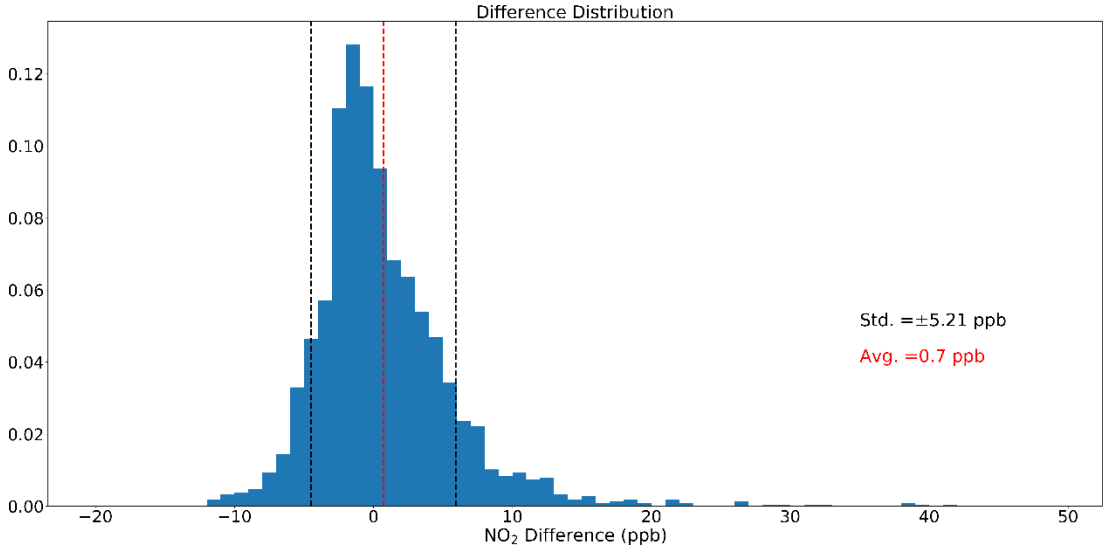
(3) Diurnal normalization: In my reading of this manuscript, the algorithm for diurnal "normalization" based on LP-DOAS measurements above the city street levels strikes me as one of the central developments presented in this manuscript, underpinning their main results. But the approach is not evaluated in any way. In my opinion, some evaluation is warranted, since it is a key aspect of their final results. I think a satisfactory evaluation could be quite easy to accomplish using the available ground-based local air quality monitoring data for which you have full diurnal observations. For example, I would propose: Why not take a random sampling of NO2 measurements at various dates and times from the stationary monitoring sites, and see if the correct diurnal profile/24-hr average can be retrieved based on the LP-DOAS normalization for that date. Repeating this experiment for, say, 50 different days from different times of day, at all three of the monitoring stations at coincident times, could quite nicely result in statistics describing how accurate this diurnal normalization approach is, depending on the time and location sampled (at least for these stationary cases). Also,

just to be clear, the output of this algorithm is essentially a “retrieved” 24-hour average concentration from a measurement at any instant in time, correct? I think this is worth spelling out very clearly. In other words, instead of calling it a “normalized” concentration, why not call it an “inferred daily average” or something like this? This would be much more intuitive. Perhaps these are not mathematically the same? (But if not, why can these numbers be compared to the WHO 24-hour average guideline concentration?)

The reviewer has correctly understood our normalization algorithm. Thanks to his/her suggestion, we replaced the description of “normalized” into “inferred daily average” when possible.

The corresponding stationary measurements from three LfU stations on mobile measurement days were used to evaluate the accuracy of the normalization algorithm. The inferred daily average concentration for each stationary measurement was retrieved based on the fitted LP DOAS normalization curve, then compared with the corresponding 24-hour average concentration measured by the same station. The distribution of the concentration differences is shown in plot below (Figure 3). The averaged difference is 0.7ppb with a 1 sigma uncertainty of 5.21ppb. The inferred daily averages compared to the actual average concentrations at the stations Landshuter Allee and Stachus differ more because due to frequent traffic jams and stop-and-go traffic they show a different diurnal cycle. Overall, the normalization algorithm effectively separated the temporal and spatial influence for most mobile measurement locations.

We added the same description in the paper (Section 3.1.1, line 15-20).



(4) Comparison of NO2 concentrations at different heights and distances: I may not have understood correctly, but is this section based on measurements from a single monitoring station over two individual days? At the beginning of Section 3.2, the authors state “measurements were conducted over two days” in March, but then later in the same section, the authors state “we repeated the 1.5 m to 4 m measurement height comparison on several days at different seasons and derived the same factor”. This is confusing and requires clarification. Please be specific regarding the deployments. As written, it seems like the authors have derived the rules for vertical and horizontal NO2 diffusion based on one specific monitoring location from two days in March, and applied that generally for their mobile results across the whole city taken in June and July. Wouldn’t the dilution and chemistry of NO2 be specific to each micro environment, where the monitoring is done? Buildings, wind patterns, light availability, I would expect to all play a role in the spatial scale of NO2 concentrations at such small scales, which would vary depending on location. Likewise, I’m surprised that these spatial scales are independent of local meteorology, and seasonality (driving O3 concentrations and other factors for example). I think this requires some evaluation, clarification, and some reasonable caveats. Finally,

isn't there prior work to cite, related to how NO2 is observed to decay by distance from, say, a highway? How do your results compare? Are the spatial scales that different?
The full analysis for NO_2 concentrations at different heights and distances has been done for 2 complete days at one station as described in the paper. Additional roadside measurements regarding the comparison between the 1.5m and 4m measurement height were conducted next to LMU Building and Landshuter Allee on several different days during different seasons and derived a similar result. Besides, the reviewer has a good point, those scaling factors depend on a lot of other factors, local meteorology, and seasonality as he/she suggested, but also local topography, driving speed and speed of the other cars, and so on. We analyzed how the uncertainty of the scaling factors contribute to the comparison with the WHO threshold shown in Table 2 and found that the percentage of exceedances can vary between 0.4% and 23.8%. It is because those dilution factors could vary, as they depend on factors in small scale environment, such as local meteorology, seasonality, local topography, urban morphology, and traffic conditions. We don't assume that the scaling factors can be applied to all our mobile measurements but rather on average for our statistical analysis. For further distance-decay studies, it is essential to carry out more extensive and long-term side-by-side measurements to cover different measurement environments (such as different locations, seasons, etc.). Modified description locates in Section 3.2, Page12, line 1-5.

(5) Comparison with OMI: It's not clear to me what is gained from the comparison with OMI in Section 3.3, and how it is relevant to this study. Essentially, the authors have demonstrated that the LP-DOAS and OMI observations share seasonality. They also conclude that the OMI-inferred values are lower than the LP-DOAS values. I don't see much new here, but perhaps the authors could clarify. How exactly does this comparison support the main conclusions of this manuscript, which involve extremely small spatial scales within a small period of time that would not be well captured by OMI?
We thought that it would be helpful to include satellite measurements, since we compare measurement methods on different scales. Point measurements from the monitoring stations and mobile measurements are combined with path averaging LP DOAs measurements, and satellite measurements provide area or volume averaging concentrations, which are more representative for a larger area, but also include vertical profiles. We agree that it is not really important for the rest of the study, so according to both reviewers' suggestion, we decided to remove it from the paper, but provide the OMI comparison as supplementary information.

(6) Main result: In my opinion, one of the main results of this manuscript is how many exceedances of the WHO guideline would actually be inferred with a more dense monitoring strategy, compared to the sparse observations currently available. But this result gets glossed over in the rest of the discussion and concluding remarks. I'd be curious if the authors agree that this one of the most important results, and if they think it deserves more prominent position in the abstract/conclusion.
We agree with the reviewer that it is one of the important results. However, we don't want to focus on the number of exceedances of the 1-hour average WHO threshold, because for the comparison we include a lot of assumptions, including the assumption that the drive-by measurement can represent the hourly average. The guideline reference value is not designed for on-road measurements. We think that the direct comparison between on-road concentrations and concentrations sampled at the monitoring stations is more representable.

The specific comments below from the reviewer also have been addressed and corrected in the manuscript.
Specific comments:
Introduction:
Page 2, Line 11: "NO2 levels are often strongly correlated with many other toxic air pollutants". Perhaps this is indeed well-known, but I think this statement merits some citations.
Relevant publications have been added as references.

Page 2, Line 25: “measures” should be “measure”
As reviewer suggested, we corrected it.

Page 2, Line 25: The authors refer to “defined rules” in terms of measurement height and distance in several places throughout the manuscript, but have mentioned what these rules are anywhere. I assume these “rules” may vary by jurisdiction. Are there consistent, e.g. European-wide, rules? Does the WHO provide location guidelines? I think at least providing the “rule” for the local agency would be insightful to share somewhere. Even better, share the rules for other places if they vary.
The European directive 2008/50/EC lists a number of criteria for microscale positioning of air quality measurements (Annex III section C). This has been added in Introduction, line 25-30.

Page 3, Lines 15-20: It seems to me that certain portions of the introduction, including these lines, would be better placed in the Methods section of this manuscript.
Since we decided to move OMI section to Supplementary, we keep the demonstration of our purpose there.

Methods:
Page 5, Line 13: The names of each urban station means very little to someone unfamiliar with the region. I later discovered these locations are indeed on the map in Figure 3c. I suggest the authors mention here that these locations can be found on this map.
As reviewer suggested, description added.

Page 6, Lines 4-11: The authors explain that the OMI VCDs are gridded onto a 0.02 x 0.02 grid following approaches from previous work, and later in the manuscript it looks like the authors use monthly averages. Can the authors be slightly more clear here? Are the observations from each day gridded separately, then I assume simple monthly averages are calculated from these daily surfaces?
Since OMI only has one measurement per day pass over Germany, and it is able to detect cloud radiance fraction, cloud pressure and albedo, too. Based on different weather condition, the viewing pixel may be missing and has large uncertainty. Each day is gridded separately and then averaged for one month. The description is added in Supplementary Section 2 line 10-15.

Page 6, Lines 13-20: Can the authors be clear about what model profiles are being used? I.e., what time of day, and I assume from the same month/year as the satellite observations? Is the surface estimate calculated each day, prior to calculating the monthly average? Or is the surface estimate performed on the monthly average columns? I believe the authors answered these questions in the response to the quick review, but why not include these details in the text for the benefit of readers? Regarding the conversion to surface concentrations, can the authors describe the logic behind using spatially interpolated profiles across four grid cells? Doesn’t this represent interpolating over an area of 4x5 degrees? I am curious why they would not simply use the spatially coincident model grid cell.
We decided to move OMI section to Supplementary.

Page 6, Lines 21-22: I’m not sure I agree entirely here. What if the diurnal pattern of sources or chemistry is different over the area of the satellite pixel, compared to the area covered by the LP-DOAS absorption path. Wouldn’t that cause a mismatch between the diurnal cycle integrated over each?
That’s a good point, we replaced “A good correlation would allow the assumption that the relative diurnal cycle obtained from the LP DOAS can be regarded as being representative for the entire urban area.” with “This correlation refers to the spatial coverage of course, and not necessarily also to the diurnal cycle, since OMI measures only once per day.”

Results and Discussion:
Page 7, Line 11: “as a general indicator”. An indicator of what?
The indicator of city-wide distribution of air quality.

Page 7, Line 13: Why have the concentrations been averaged over the three locations in this case, which presumably represent quite different micro environments? Why not just report the average concentration of each separately, and within each of their 10-km radius? Section 3.1.1: As I mention above, this really strikes me as belonging in the Methods section. This means you could also keep Figure 2 before Figure 3. As it is now, you reference Figure 3 in the text before discussing Figure 2.
Answered in General comments (2).

Figure 3c: You could be very clear here that this is a zoomed-in plot of the diurnally normalized data.
Yes. It is now clearly indicated in the plot caption.

Table 1: You could be very clear here that the WHO AQG is according to the 24-hr average guideline.
The caption of Table 2 is modified to WHO AQG 1-hour guideline.

Page 10, Line 17: This is assuming that the WHO guidelines are based on the same monitoring “rules” that are applied by the environmental reporting agency. Is this true? What are the WHO guidelines for monitor placement? Are these the same rules employed by the local air quality agency? Please share these rules explicitly for the benefit of the readers.
Here, according to the European directive 2008/50/EC, a number of criteria for microscale positioning of air quality measurements (Annex III section C) are introduced. This has been added in Introduction, line 25-30.

Reply to Anonymous Referee #3

1) The mobile data set is interesting. However other mobile data sets examining spatial variability in cities exist and it is not clear if any of the observations here are surprising because they are not placed in the context of the prior related literature. I recommend adding a deeper and more comprehensive discussion of the theoretical and observational understanding we have of emissions from roadways and the length scales of decay of those emissions in cities. For example, papers by Choi et al. including https://doi.org/10.1016/j.atmosenv.2012.07.084 and Atmos. Chem. Phys., 14, 6925–6940, 2014 and by Apte et al. Environ. Sci. Technol. 2017, 51, 12, 6999–7008 show a characteristic decay length scale of 500-1000m (1/e) that would be approximately consistent with the measurements reported herein. The Choi et al papers also provide a theoretical basis for discussion of the decay. It is also important to note that the time scale for conversion of NO to NO2 is not instantaneous. Thus on-road measurements of NO2 may have a systematic bias. The measurements in Apte, et al. show the consequences of NO to NO2 conversion as different timescales/lengthscales for decay from urban roadway sources.

Thanks very much for reviewer's suggestions. All publications above have been added as references in Section 3.2 line 15-20. These are valuable information to support our research.

2) I find the discussion of the remote sensing measurements confusing. The logic connecting them to the mobile measurements is unclear. It is well-known that OMI measurements with a 2 degree a priori will have a large bias compared to urban measurements. The large context of the long path measurements connects both emissions and loss, while the mobile measurements are so near to the source that they only reflect emissions. I recommend these sections be removed or the connection to the mobile observations made substantially clearer. Also, to help with the readability of the paper, I recommend moving all descriptions of the instruments to the supplement.

According to the reviewer's suggestion, we decided to remove the whole section of OMI comparison from the paper and move it to the supplement

The description of the instruments is kept in the paper, because we think it is important to describe the different measurement techniques for providing a better understanding for the combination of point and path averaging measurements.

List of All Relevant Changes

1. Page 2
    a. Line 11:
    Add new references.
    b. Line 25 - 27:
    Change "Relative regulations" into “Related regulations”.
    Add EU regulation regarding the position of air quality measurements.
2. Page 3
    a. All change contents:
    Move all relevant OMI satellite description into the Supplement, meanwhile, add a brief summary about the OMI satellite observation.
3. Page 4
    a. Line 15 - 20:
    Add more detailed information about measurement days and the resolution of NO_2 map.
4. Page 5
    a. Line 0:
    Add Table 1: Overview of the measurement days, time and number of measurements.
    b. Line 11 - 15:
    Description about the light paths of LP DOAS was added.
5. Page 6
    a. Line 8:
    Add the description ‘locations of the three stations are also presented in’ for Fig.4c.
6. Page 7
    a. All change contents:
    Move all relevant OMI satellite description into the Supplement.
7. Page 9
    a. Line 5 - 13:
    Add the evaluation of the accuracy of the normalization algorithm.
8. Page 11
    a. Line 3 - 5
    Add ‘Note that WHO reference concentration refers to 1h average values, whereas we measure at each location for a few seconds. We use this comparison to show how often this threshold would be exceeded if the drive-by measurement were representative local respective situations.’
9. Page 12
    a. Line 0
    Add ‘1-hour guideline’ in the caption of Table 2.
    b. Line 9 – 11
    Add new references about the distance decay studies.
    c. Line 15 - 19
    Modified the description.
    d. Line 20 - 24
    Add the analysis about the varying percentages of the exceedances of WHO 1-hour guideline.
10. Page 13
    a. Line 0
    Add ‘with a standard deviation of 0.18’.
    b. Line 9 - End of Section 3.3
    Move all relevant OMI satellite observation study into the Supplement.

Spatial and temporal representativeness of point measurements for nitrogen dioxide pollution levels in cities

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Abstract. In many cities around the world the overall air quality is improving, but at the same time nitrogen dioxide (NO_2) trends show stagnating values and in many cases could not be reduced below air quality standards recommended by the World Health Organization (WHO). Many large cities have built monitoring stations to continuously measure different air pollutants. While most stations follow defined rules in terms of measurement height and distance to traffic emissions, the question remains, how representative are those point measurements for the city-wide air quality. The question of the spatial coverage of a point measurement is important because it defines the area of influence and coverage of monitoring networks, determines how to assimilate monitoring data into model simulations or compare to satellite data with a coarser resolution, and is essential to assess the impact of the acquired data on public health.

In order to answer this question, we combined different measurement data sets consisting of path averaging remote sensing data and in-situ point measurements in stationary and mobile setups from a measurement campaign that took place in Munich, Germany in June and July 2016. We developed an algorithm to strip temporal diversity and spatial patterns, in order to construct a consistent NO_2 pollution map for Munich. Continuous long-path differential optical absorption spectroscopy (LP DOAS) measurements were complemented with mobile cavity-enhanced (CE) DOAS, chemiluminescence (CL) and cavity attenuated phase shift (CAPS) instruments and were compared to monitoring stations and satellite data. In order to generate a consistent composite map, the LP DOAS diurnal cycle has been used to normalize for the time of the day dependency of the source patterns, so that spatial and temporal patterns can be analyzed separately. The resulting concentration map visualizes pollution hot spots at traffic junctions and tunnel exits in Munich, providing insights into the strong spatial variations. On the other hand, this database is beneficial to the urban planning and the design of control measures of environment pollution. Directly comparing on-street mobile measurements in the vicinity of monitoring stations resulted in a difference of $48\,\%$. For the extrapolation of the monitoring station data to street level, we determined the influence of the measuring height and distance to the street. We found that a measuring height of $4\,\mathrm{m}$, at which the Munich monitoring stations measure, results in $16\,\%$ lower average concentrations than a measuring height of $1.5\,\mathrm{m}$, which is the height of the inlet of our mobile measurements and a typical pedestrian breathing height. The horizontal distance of most stations to the center of the street of about $6\,\mathrm{m}$ also results in an average reduction of $13\,\%$ compared to street level concentration. A difference of $21\,\%$ in the NO_2 concentrations

remained, which could be an indication that city-wide measurements are needed for capturing the full range and variability of concentrations for assessing pollutant exposure and air quality in cities.

1 Introduction

Many former studies [*Huang et al.*, 2014; *Dunlea et al.*, 2007; *Jang and Kamens*, 2001] have been pointed out that NO_2 is an important composition in the process of both tropospheric and stratospheric chemistry. It is one of the major pollution products from combustion processes. Catalytic formation of tropospheric ozone (O_3) and the formation of secondary aerosols that cause acid rain, all of which involve its participation. Elevated concentration of atmospheric NO_2 is acknowledged to be noxious to human beings health. In urban environments, Exhaust emissions are one of the primary sources of air pollution, particularly NO_x (= NO + NO_2). Nitrogen-monoxide (NO) accounts for the majority of direct traffic emissions, which is subsequently oxidized to form NO_2 although some NO_2 is emitted directly [*Ban-Weiss et al.*, 2008; *Henderson et al.*, 2007; *Kirchstetter et al.*, 1999]. NO_2 levels are often strongly correlated with many other toxic air pollutants [*Massman*, 1998; *Yoo et* al., 2014; *Xie et* al., 2015]. Its concentration can be easily and precisely measured, which is helpful in assessing general air quality. Since it is a short-lived compound gas from numerous different sources, its concentrations can vary strongly, both in space and time.

According to the 2017 European Environment Agency report [*EEA*, 2017], some NO_2 concentrations measured at air quality monitoring stations are above the World Health Organization (WHO) Air Quality Guideline (AQG) values of $200\,\mu g/m^3$ (hourly) and $40\,\mu g/m^3$ (annually). $10.5\,\%$ of the stations across European cities exceeded the annual limits including several German cities. None of the exceedances were observed at rural background stations, but in urban or suburban stations. More specifically, $89\,\%$ of the exceeded values were observed at traffic stations. The 2016 air quality report by the German Environment Agency (Umweltbundesamt) [*UBA*, 2017] also pointed out that the air pollution in urban conurbations was primarily affected by traffic. In the 2015 technical report by the Bavarian environment agency (Landesamt für Umwelt, LfU) [*LfU*, 2015], the on-road NO_2 concentration limits were exceeded in most Bavarian cities from 2000 to 2014. In particular, the annual NO_2 level in Munich measured at Landshuter Allee station was more than twice of the annual NO_2 limit value of the WHO AQG of $40\,\mu g/m^3$.

With a growing focus on air pollution in the public attention, stationary monitoring networks have been established all over the world. ~~Monitoring stations continuously measures~~ Related regulations also have been certified. For example, in Europe, the European directive 2008/50/EC lists a number of criteria for micro-scale positioning of air quality measurements (Annex III section C). Monitoring stations in EU continuously measure different pollutants and while most stations follow the defined rules in terms of measurement height and distance to traffic emissions, the question remains, how representative are those point measurements for the city wide air quality. According to a study of the spatial distribution of NO_2 in Hong Kong [*Zhu et al.*, 2018], large differences between mobile measurements around the city and seven local monitoring stations were observed. In order to determine the representativeness of air quality monitoring stations, different measurement methods have to be combined. Most monitoring stations utilize the ChemiLuminescence (CL) technique for NO_x measurements. Thereby the NO_2 concentration is determined indirectly by calculating the difference between NO_x and NO concentrations. The concentration

of oxidized odd-Nitrogen species (NO_y) is inevitably included as a small measurement error. Nevertheless, the CL technique has a good detection sensitivity that is given by its low background signal. This is because for initiating the fluorescence no light source is required [*Dunlea et al.*, 2007]. In this study, we compared our CL and cavity-enhanced DOAS (CE DOAS) data to the local air quality stations and studied the diffusion rate of NO_2 in both vertical and horizontal directions from one of the stations.

For our study we utilized a combination of long-path DOAS (LP-DOAS) instrument and a CE DOAS, as well as a Cavity Attenuated Phase shift Spectroscopy (CAPS) instrument to determine the spatio-temporal variability of NO_2 concentrations in the central area of Munich, CE DOAS is a spectroscopic measurement technique that uses an optical resonator to fold the absorption path into into a resonator [*Zhu et al.*, 2018; *Min et al.*, 2016; *Thalman and Volkamer*, 2010; *Platt et al.*, 2009; *Washenfelder et al.*, 2008; *Venables et al.*, 2006; *Langridge et al.*, 2006]. CAPS [*Herbelin et al.*, 1980] is a spectroscopic detection technology, generally referred to cavity enhanced optical absorption, which has also been applied for the detection of atmospheric pollutants in many studies [*Xie et al.*, 2019; *Kundu et al.*, 2019; *Ge et al.*, 2013; *Kebabian et al.*, 2008, 2005a]. The advantage of CE DOAS and CAPS is the fact that they are not sensitive to other reactive nitrogen oxides in the atmosphere like some other in-situ NO_2 monitoring techniques. They are both characterized by a compact setup and have no sensitivity loss during the operation. For mobile measurements a fast sampling rate is necessary, and the high accuracy of the instruments allowed a sampling rate of 2 s. Similar instrument setups have been used in many on-road studies of vehicles emissions [*Zhu et al.*, 2018; *Chan et al.*, 2017; *Rakowska et al.*, 2014; *Ning et al.*, 2012; *Uhrner et al.*, 2007; *Vogt et al.*, 2003].

~~In order to verify whether the LP DOAS measurements are representative for the whole city, NO_2 data from the Ozone Monitoring Instrument (OMI) on-board the NASA Aura satellite was used. Satellite measurements are commonly used for global scale long-term observation of aerosols and trace gases *Silvern et* al., 2019; *Zara et* al., 2018; *Laughner and Cohen*, 2017; *Inness et* al., 2015. Validation studies revealed that satellite retrievals generally underestimate urban areas but also found good correlations between satellite and ground based observations *Chan et* al., 2018; *Lin et* al., 2012; *Lamsal et* al., 2008; *Wenig et* al., 2008; *Petritoli et* al., 2004.~~

For our study we conducted on-road measurements of NO_2 concentrations in June and July of 2016 in order to investigate street level air quality and locate emission hot spot areas. Additionally, LP DOAS measurements were conducted to observe the temporal variability of ambient NO_2 in Munich. A measurement system consisting of several DOAS instruments was continuously operational for over 2 years (see Sec. 2). The algorithm that combines the mobile and stationary measurement data is described in Section 3.1.1. The resulting on-road NO_2 spatial patterns are presented in Section 3.1.2. The CE DOAS and CL were set up next to the Bavarian LfU local air quality station to measure the horizontal and vertical NO_2 distributions, and the results are shown in Section 3.2. ~~In addition, Section ?? presents the comparison of LP DOAS~~ NO_2 ~~measurements with OMIsatellite data, and analyzes the characteristics of seasonal NO_2 variation.~~ data from the Ozone Monitoring Instrument (OMI) on-board the NASA Aura satellite was used to verify how the LP DOAS measurements compare to the much larger OMI ground pixels covering a larger fraction of the city in order to find out whether the LP DOAS measurements are representative for the whole city. Detailed analysis is provided in the supplementary document.

2 Methodology

This study combines different measurement methods such as mobile, stationary and satellite measurements to answer the question of how representative sparse point measurements are to determine the air quality of a city. Furthermore, we want to find out what kind of measurement approach is needed to determine the overall air quality in a city. The Munich three-dimensional DOAS measuring system combines three different types of DOAS instruments, specifically, CE DOAS and LP DOAS. The measurement system is installed on the roof of the building of the Meteorological Institute Munich (MIM) at the Ludwig Maximilians University (LMU) in the center of Munich. The three LP DOAS instruments scan retro reflector arrays in different directions and distances, capturing the horizontal variations at the rooftop level. A CE DOAS is used to determine the NO_2 variability on the ground. The LP DOAS instruments run continuously, whereas the CE DOAS is used at different times of the year/week/day under varying meteorological conditions to determine street-level NO_2 distributions.

2.1 Mobile Measurements

A CE DOAS and a CAPS instruments in two vehicles were used for on-street sampling of traffic emissions. The sampling inlets were located at the front right window of each vehicle at $1.5\,\mathrm{m}$ height. For measurements in Munich's city park (English Garden), we used a bike trailer. The measurements were performed ~~in June and~~ on two days in March, three days in May, five days in June and six days in July 2016 to cover a large part of the urban area in Munich. Table 1 shows the dates, measurement times and number of measurements for all measurement days. The sample resolution of the CE DOAS and the CAPS were both adjusted to $2\,\mathrm{s}$ during the mobile measurements. The measurements were performed on varying routes during daytime to cover the entire city center area. All measurements were spatially averaged to a high-resolution grid of $20\,\mathrm{m} \times 20\,\mathrm{m}$. This resolution has been chosen according to an average driving speed of $40\,\mathrm{km\,h^{-1}}$ to $50\,\mathrm{km\,h^{-1}}$ and the measurement sampling time of $2\,\mathrm{s}$, so that at least one measurement value per drive-by falls into each grid box. All measurements from different drives and different days are averaged for each grid cell after diurnal cycle normalization.

The CE DOAS is composed of an air sampling system, an optical resonator with two high reflective mirrors, a blue LED light source and a spectrometer [*Platt et al.*, 2009]. For the spectral retrieval in the wavelength range $435.6\,\mathrm{nm}$ to $455.1\,\mathrm{nm}$, we used DOASIS [*Kraus*, 2005]. The NO_2 reference absorption cross is from *Vandaele et al.* [2002], O_4 from *Hermans et al.* [1999], H_2O from *Rothman et al.* [2003] and CHOCHO (Glyoxal) from *Volkamer et al.* [2005].

The CAPS measurement technique is closely related to Cavity Ring-Down Laser absorption Spectroscopy (CRDS), which determines the concentration of trace gases from the decay rate of the light source in the optical resonator [*Ball and Jones*, 2003; *Brown et al.*, 2002; *Berden et al.*, 2000; *Engeln et al.*, 1996]. CRDS is a laser-based system, while CAPS uses an incoherent light source (a blue LED) that is well-matched to the NO_2 absorption band. The CAPS NO_2 system mainly consists of a blue LED, a measurement chamber with two highly reflective mirrors centered at $450\,\mathrm{nm}$, and a vacuum photodiode detector. It estimates the NO_2 concentration by directly measuring the optical absorption of NO_2 at the $450\,\mathrm{nm}$ wavelength within the electromagnetic spectrum. The light appears as a distorted waveform after passing through two mirrors and the measurement cell, which is characterized by a phase shift that is determined by demodulation techniques in comparison to the initial LED

Table 1. Overview of the measurement days, time and number of measurements.

Meas. Date	Meas. Time (hours)	Number of Meas.	Meas. Date	Meas. Time (hours)	Number of Meas.
2016.03.21	3.84	5313	2016.06.16	5.48	7586
2016.03.22	1.82	2523	2016.06.17	2.75	3804
2016.05.09	2.29	3172	2016.07.01	2.86	3955
2016.05.10	3.62	5019	2016.07.06	6.24	17292
2016.05.11	4.55	6295	2016.07.07	10.29	28489
2016.06.06	3.38	4674	2016.07.08	5.22	14466
2016.06.07	4.66	6454	2016.07.11	5.1	14123
2016.06.13	4.25	5882	2016.07.13	2.48	2976
			Total	64.8	126440

light modulation. The phase shift is proportional to the absorbance of the light by the presence of NO_2. The concentration of NO_2 can be derived by measuring the amount of the phase shift. The detailed principles of the CAPS system are demonstrated in *Kebabian et al.* [2008, 2005b].

2.2 Long-path (LP) DOAS observations

Three LP DOAS instruments were installed on the roof of the MIM. The measurement setups are displayed in Figure 1. The measurement system started operation in December 2015 with a total absorption path of $3828\,\mathrm{m}$ across the English Garden to a retro reflector array located on the rooftop of the Hilton hotel building at $\sim 48\,\mathrm{m}$ height. In January 2017 another absorption path of $1142\,\mathrm{m}$ was installed covering three blocks around the university area to a retro reflector at the St. Ludwig Munich Church at $\sim 40\,\mathrm{m}$ above ground. Since July 2015 a retro reflector is also installed at the roof of the N5 building of the Technical University of Munich (TUM) at $\sim 28\,\mathrm{m}$ height, allowing an absorption path of $828\,\mathrm{m}$. From July 2016 to August 2017 a path of $816\,\mathrm{m}$ to the roof of the building of the Physics department of LMU at $\sim 24\,\mathrm{m}$ height was operational as well. For the long-term average diurnal cycle calculation described in Section 3.1.1 all available data from all measurement paths were used. LP DOAS measurements of light paths towards to the Hilton hotel, TUM University, and LMU physic department were used to normalize the mobile measurements regarding the diurnal cycle. The measurement paths cover the university campus, the public park, residential areas and areas with heavy traffic. The instrumental background was corrected by subtracting the LED reference spectra, including dark current, offset, and background, from each measured spectrum.

A measurement sequence starts by taking a LED reference spectra using a shortcut system consisting of a diffuser plate in front of the y-fiber and an exposure time of $10\,\mathrm{s}$. Then a shutter is used to block the LED for measuring the atmospheric

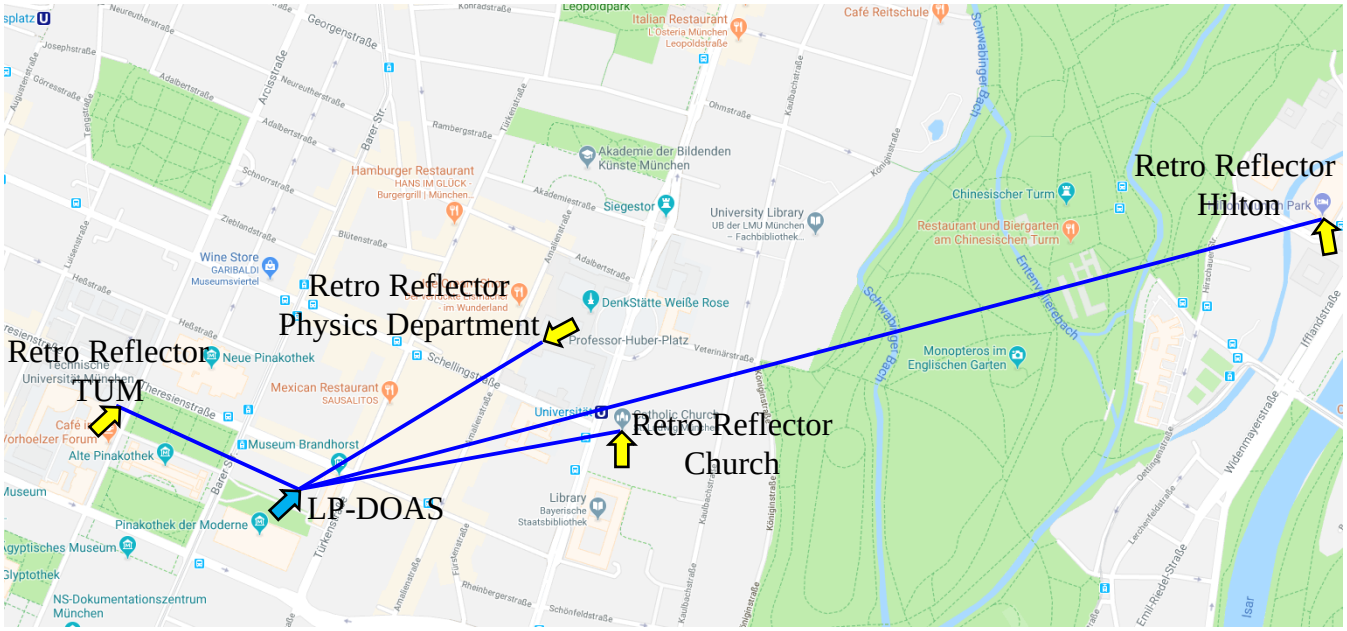

Figure 1. Map of Munich city center and four optical paths of three LP DOAS instruments. Map data ©Google maps.

background spectrum for 1 s. Afterwards, the atmospheric spectrum with a maximum of 10 scans is taken. Each scan of a spectrum has a peak intensity of about 60 % to 80 % saturation of the detector and typically requires 60 ms to 1000 ms, depending on the visibility and instrument setup. The total sampling time (the product of the number of scans and exposure time for each scan) was limited to 60 s. A full measurement sequence took between 30 s and 90 s, depending on visibility conditions.

2.3 Local air quality monitoring network

The Bavarian LfU is operating five monitoring stations, three roadside stations at Landshuter Allee, Stachus and Lothstrasse (locations of the three stations are also presented in Figure 4c), and two ambient stations in Allach and Johanneskirchen. In these stations the air pollutants NO, NO_2, CO, O_3, $PM_{2.5}$, PM_{10} and in addition meteorological parameters such as relative humidity and temperature are measured. In this study we concentrated on the NO and NO_2 concentrations that are continuously monitored using an in situ CL NO_x analyzer (HORIBA APNA-370) [*LfU*, 2019].

2.4 ~~The Ozone Monitoring Instrument (OMI) satellite observations~~

~~The Ozone Monitoring Instrument (OMI) is an imaging spectrometer on board the NASA Aura satellite. It measures earthshine radiances with two grating spectrometers which cover the wavelength range from 264 nm to 504 nm. OMI provides the daily~~

measurements of NO_2, BrO, SO_2, O_3, HCHO, OClO, BrO and aerosols in a global coverage. It is able to detect the cloud radiance fraction, cloud pressure and albedo.

NASA's OMI standard product version 3 (SPv3) *Krotkov et* al., 2017; *Marchenko et* al., 2015is used in this study. The OMI NO_2 data is publicly available at the Goddard Earth Sciences Data and Information Services Center (GES DISC) (https://disc.gsfc.nasa.gov/). For our comparison we gridded the OMI VCDs onto a high resolution grid with $0.02° \times 0.02°$ following the approach described in previous studies *Chan et* al., 2015; *Kuhlmann et* al., 2014.

2.3.1 Converting OMI vertical column densities to ground mixing ratio using modeled NO_2 profiles

In order to examine how representative the LP DOAS data is for the temporal pattern observed by OMI, which covers the entire city, OMI's vertical column densities (VCDs) are converted into ground concentrations. For the conversion, vertical profile information is needed. We utilized NO_2 vertical profile information simulated by the chemistry transport model (CTM) GEOS-Chem *Bey et* al., 2001. The horizontal resolution of the simulation is $2.0°$ (latitude) $\times$ $2.5°$. Vertical profiles of NO_2 are spatially interpolated within the 4 closest grid cell to the measurement location. Detailed description of the GEOS-Chem simulation can be found in previous studies *Chan*, 2017a, b.

Since we use the LP DOAS data for the diurnal correction, we tested the correlation between LP DOAS measurement values and OMI data. The OMI satellite measurements cover a larger area of Munich with the instrument's ground pixel footprint of $\sim$320 - 6400 km^2 than the 2 km path length of the LP DOAS instrument. A good correlation would allow the assumption that the relative diurnal cycle obtained from the LP DOAS can be regarded as being representative for the entire urban area.

3 Results and Discussion

3.1 NO_2 concentration maps constructed using mobile measurements

The mobile measurement data can be used to create a map showing the city-wide distribution of air quality using NO_2 concentrations as a general indicator (Figure 4). As a first test, we compared the averaged measurement values within a $10\,\mathrm{km}$ radius around the three governmental monitoring stations at Landshuter Allee, Lothstrasse and Stachus and obtained an averaged concentration of $93\,\mu\mathrm{g/m^3}$ for the mobile measurements and $48\,\mu\mathrm{g/m^3}$ for the three stations for the campaign days in June and July 2016. The large difference can be explained by looking at the criteria for the location of monitoring sites set by the European Union: the recommended measurement height is between $1.5\,\mathrm{m}$ and $4\,\mathrm{m}$, maximum distance to the street is $10\,\mathrm{m}$ and at a minimum distance to the next crossroad of $25\,\mathrm{m}$ [*Commission*, 2008]. Most monitoring stations have the inlet positioned at $4\,\mathrm{m}$ height. The mobile measurement data, however, include the significantly increased concentrations at crossroads, tunnel exits and other pollution hot spots. In addition, the height of the measurement inlets differs by $2.5\,\mathrm{m}$ between the mobile measurements and the governmental monitoring stations, which also influences the comparison. In order to determine how representative point measurements are for the city-wide air quality, we analyzed the correlation between point measurements and the distribution captured by mobile measurements, between point and path averaging measurements, and between path av-

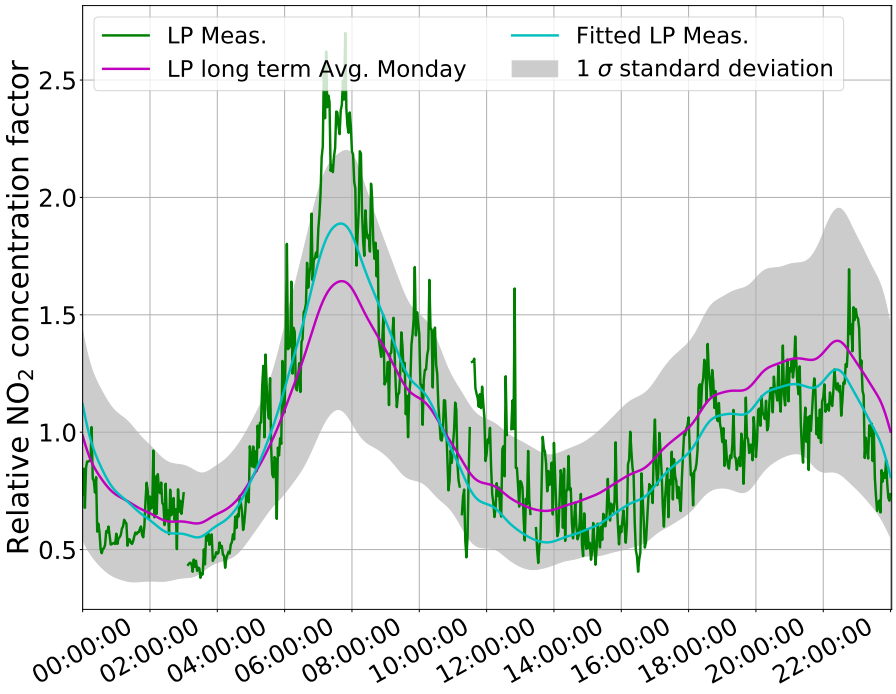

Figure 2. Normalization curve used to correct mobile measurement data. The purple curve is the long term average diurnal pattern for the day of the week of the measurement day (Monday in this example), which is fitted (scaling with linear time dependent factor and offset) to the measurement data of 13^{th} June 2016 shown in green, excluding the data outside of the $2\,\sigma$ area shown in gray. The resulting cyan curve is used to remove the diurnal dependency of the mobile measurements data.

eraging and satellite measurements. Since the spatial distribution can not be captured instantaneous, an algorithm to normalize for the diurnal variation is needed in order to create a consistent map representing only the spatial variability of daily average concentrations instead of temporal influences.

3.1.1 Normalization of the diurnal cycle

As a mobile survey cannot capture the concentrations at different locations simultaneously, and the NO_2 measurements are naturally influenced by daily variations such as changing boundary layer height or the diurnal cycle of the traffic amount, we use an algorithm to separate temporal and spatial patterns in the data set.

First, the algorithm normalizes the long time series of LP DOAS measurements of atmospheric NO_2 by dividing through the daily average NO_2 concentration of the same day. The mean concentration curves for each day of the week over a period of 2.5 years are calculated in order to obtain a relative diurnal NO_2 variation pattern (purple curve in Figure 2). The normalized averaged diurnal NO_2 curve of the corresponding weekday is fitted (using an offset and a scaling with a linearly time dependent factor) to the normalized LP DOAS measurement of the corresponding day, coinciding with the mobile measurements. In order

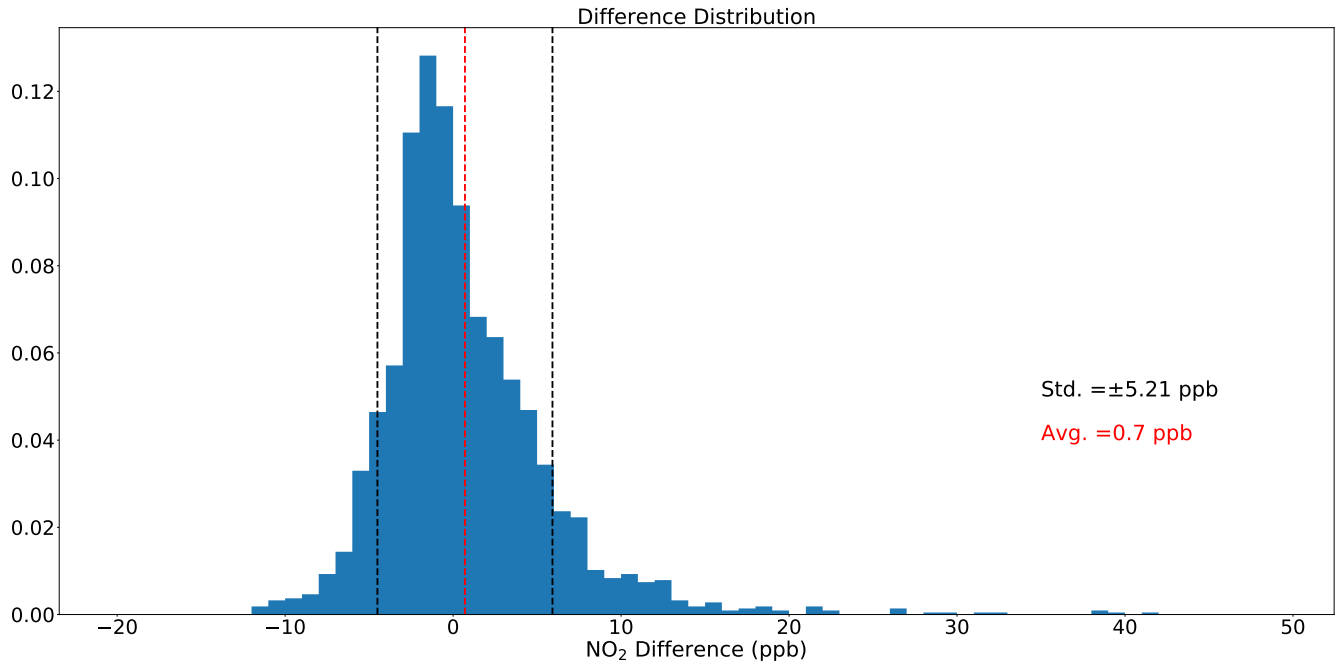

Figure 3. The distribution of the differences in between of inferred daily average concentrations and 24-hour average concentrations measured by LfU stations. Bin width is 1 ppb. Evaluated stations including Allach, Johanneskirch and Lothstraße.

to remove the influence of outliers, NO_2 values outside of ~~2σ~~ $2\,\sigma$ variation of the fitted curve are disregarded (cyan curve). Figure 2 shows the fitting process for the normalization curve for one day of the measurement campaign. The other days show very similar characteristics with a significant peak in the morning and evening rush hours. Dividing the mobile measurement data by the curve data, removes the diurnal dependencies and allows focusing on spatial pattern.

The corresponding stationary measurements from three LfU stations on mobile measurement days were used to evaluate the accuracy of the normalization algorithm. The inferred daily average concentration for each stationary measurement was retrieved based on the fitted LP DOAS normalization curve, then compared with the corresponding 24-hour average concentration measured by the same station. The distribution of the concentration differences is shown in Figure 3. The averaged difference is 0.7 ppb with $1\,\sigma$ uncertainty of 5.21 ppb. The inferred daily averages compared to the actual average concentrations at the stations Landshuter Allee and Stachus differ more because due to frequent traffic jams and stop-and-go traffic they show a different diurnal cycle. Overall, the normalization algorithm effectively separated the temporal and spatial influence for most mobile measurement locations.

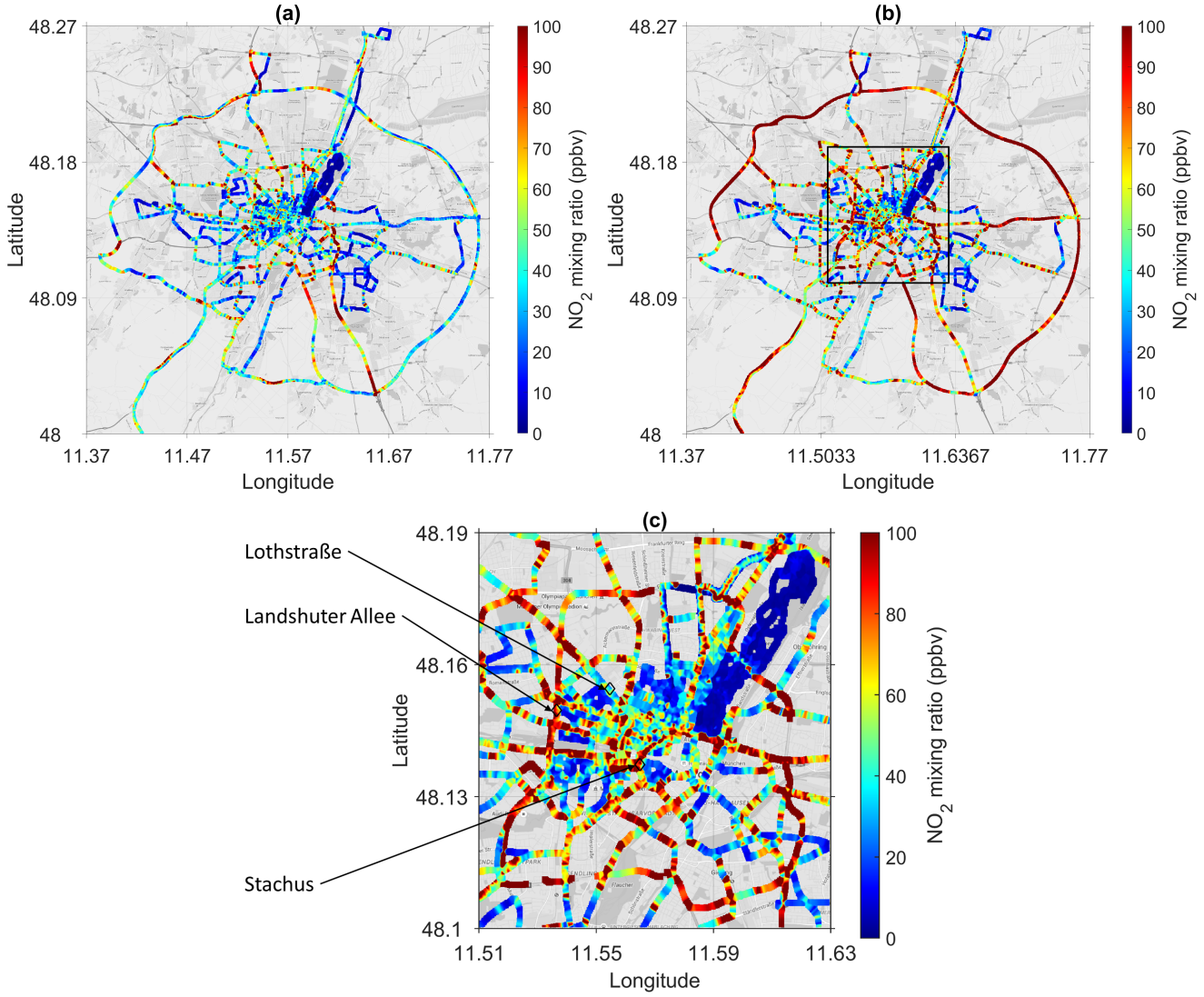

Figure 4. (a) CE DOAS and CAPS mobile measurements of NO_2 in Munich in 2016. (b) Normalized spatial distribution of NO_2 using coinciding LP DOAS data to remove the diurnal dependencies. (c) Zoom in of the city center. The three black diamonds in (c) are the locations of the governmental monitoring stations (Landshuter Allee, Lothstraße, Stachus). The area at the top right with very low concentrations represents the city park English Garden. Map data ©Google maps.

3.1.2 Spatial distribution of NO_2 in the city of Munich

The measured concentrations during the campaign were spatially averaged to a high resolution grid of $20\,\mathrm{m} \times 20\,\mathrm{m}$ (Figure 4a). Most of these measurements are distributed on major roads, including city, urban ring-road, suburbs, rural areas, and highways. Relatively high NO_2 pollution could be observed on motorways and busy urban roads. Difference between main

roads and adjoining side roads of up to a factor of 5 can be observed. $4.4\,\%$ of the on-road measurements exceeded the WHO 1-hour guideline value of $200\,\mu g/m^3 \approx 106\,ppb$ (depending on temperature, here the appropriate conversion factor at $25\,°C$ and $1013\,hPa$ are used), corresponding to ~~$6.6\,\%$~~ $4.1\,\%$ of the area covered. Note that WHO reference concentration refers to 1h average values, whereas we measure at each location for a few seconds. We use this comparison to show how often this threshold would be exceeded if the drive-by measurement were representative local respective situations. High NO_2 values over motorways were mainly due to the emission of heavy duty diesel vehicles, i.e. a significant increase could be observed when we were driving behind trucks and buses. In the city center traffic congestion and the street canyon effect [*Rakowska et al.*, 2014] can be the main cause of elevated on-road NO_2 levels. *Zhu et al.* [2018] showed in a study in Hong Kong that average pollution exposure increases by $14.5\,\%$ when stopping at a traffic light compared to fluent traffic. Other studies showed as well that the distribution of pollutants is mainly impacted by traffic flow patterns [*Fu et al.*, 2017; *Rakowska et al.*, 2014; *Huan and Kebin*, 2012; *Kaur et al.*, 2007; *Westerdahl et al.*, 2005]. The normalization using coinciding LP DOAS measurement removes the diurnal dependency but leaves the traffic flow dependency in the data, because it contributes to the city-wide air quality. The normalized on-road NO_2 map shown in Figure 4(b) represents daily average values for all locations. After normalization, there exist some regions, where NO_2 concentrations are consistently higher, while in other areas the normalized concentrations are lower than the original measurements. This behavior can be explained by the time of the day when the measurements were taken: when we measured during the rush hour, the measurements are higher, while the measurements during noon are lower than the daily average. The normalization procedure increased the occurrences of WHO 1-hour guideline exceedances to $14.5\,\%$ of the on-road measurements, corresponding to $17.1\,\%$ of the total area (including motorways) and $15.7\,\%$ of the area in the city center. However, the thresholds in WHO AQG are based on studies involving monitoring station data which are not measuring directly on the street. Taking the vertical and horizontal dilution factors (see Figure 5, the factor 0.84 for $4\,m$ height and 0.87 for $6\,m$ distance) into account, we extrapolate WHO AQG 1-hour threshold value of $200\,\mu g/m^3$ to the on-road level with the value of $273.7\,\mu g/m^3 \approx 145.6\,ppb$ ($200\,\mu g/m^3/0.84/0.87$)), then also calculated the frequency of exceedances (cf. table 2).

It can be seen that especially in the downtown area (Figure 4c), the values after the normalization are noticeable higher than before. This can be explained as we tried to avoid the rush hours, i.e. traffic jams, for performing the measurements. Therefore, the measured NO_2 level is often lower in comparison to the day-average. The area with significantly lower concentrations seen in Figure 4 is the city park (English Garden) with no vehicle emissions and where plants could provide deposition areas for O_3 , NO_x and particles [*Chaparro-Suarez et al.*, 2011; *Wesely and Hicks*, 2000].

3.2 Comparison of NO_2 concentrations at different heights and distances from the street

In order to investigate the diffusion effects of emitted NO_2 molecules in both vertical and horizontal directions, measurements were conducted over two days (20th. and 22th. March 2019) using CE DOAS and CL instruments at Stachus, Munich, next to the governmental monitoring station. Since we used two different measurement techniques, the first step was to check the instrument for consistency. Side-by-side measurements next to the street (same height and distance to the street in Figure 5) were used to analyze differences. We found the CL NO_2 to be $2\,\%$ higher, possible due to sensitivities of the molybdenum

Table 2. Percentage of measured concentrations exceeding the WHO AQG 1-hour guideline and its corrected on-road level thresholds for both temporal and spatial coverage. The values are broken down for before and after the normalization of the data according to diurnal patterns, and also calculated for WHO guideline values adjusted for the different measurement height and distance to the street.

	Percentage exceeding WHO guideline		Percentage exceeding adjusted WHO guideline	
	Before normalization	After normalization	Before normalization	After normalization
Temporally	4.4 %	14.5 %	1.7 %	6.7 %
Spatially (total)	4.1 %	17.1 %	1.1 %	4.6 %
Spatially (downtown)	5.5 %	12.4 %	1.6 %	4.7 %

oxide converters to NO_y species [see *Villena et al.*, 2012; *Dunlea et al.*, 2007]. We corrected the CL measurement data in order to remove those interferences.

Both instruments were set up next to the governmental monitoring station at Stachus, which is at a height of 4 m and has a 30 min time resolution. The CE DOAS was set up next to the street and measured at a fixed height of 1.5 m above the ground, while the CL instrument measured NO_2 at multiple heights above the ground (from 0.5 m to 4 m) and at different distances from the side of the street (from 2 m to 10 m). The temporal resolution for both instruments was set to 5 s. All measurements are shown in Figure 5(a) for the different measurement heights and (b) for the different distances to the side of the street. Figure 5(c) shows the distribution of the ratios, and it can clearly be seen that the average concentrations decrease with height and distance. Another mobile measurement study by *Apte et* al. [2017] also demonstrates the distance-decay characteristic of urban spatial distribution of NO, NO_2, and black carbon but on larger scales. More relevant theoretical decay researches can be found in *Choi et al.* [2012, 2014]. Figure 5(d) shows a two-day comparison between the 30 min average NO_2 concentrations measured with our CE DOAS at a height of 1.5 m with the CL instrument data of the governmental monitoring station at a height of 4 m. The regression plot shows a ratio of 1.23 between the measurements at 1.5 m and 4 m height. ~~We repeated the 1.5 m to 4 m measurement height comparison on several different days at different seasons and derived the same~~ In addition, the side by side measurement derives a factor of 0.84 with a standard deviation of ~~0.21~~0.20, so most ratios vary from 37 % decrease with increasing height (factor 0.84~~-0.21~~-0.20=~~0.63~~0.64) to 5 % increase (factor 0.84+~~0.21~~)0.20=~~1.05).~~ 1.04). Additional roadside measurements regarding the comparison between the 1.5 m and 4 m measurement height were conducted at roadside next to the MIM building and the LfU station of Landshuter Allee on several different days during different seasons and derived a similar result. Since the inlet height for our mobile measurements is 1.5 m, we take this factor into account when comparing to monitoring station data. In addition, we analyzed how the uncertainty of the scaling factors (horizontal and vertical) contribute to the comparison with the WHO threshold shown in Table 2 and found that the percentage of exceedances can vary between 0.4 % and 23.8 %. It is because those dilution factors could vary, as they depend on factors in small scale environment, such as local meteorology, seasonality, local topography, urban morphology, and traffic conditions. We don't assume that the scaling factors can be applied to all our mobile measurements but rather on average for our statistical analysis. For further

distance-decay studies, it is essential to carry out more extensive and long-term side-by-side measurements to cover different measurement environments (such as different locations, seasons, etc.). In terms of distance to the street, measuring at the center of the street, like we did during the mobile measurements, and measuring at a distance of 6 m, which is approximately the distance of most monitoring stations to the middle of the street, the on-road measurements are 13 % higher with a standard deviation of 0.18 due to the observed diffusion effects. Those factors have to be kept in mind when comparing on-road measurements to monitoring station data or any other measurement data taken at different height levels and distances to the street. This leads to the conclusion, that from the 48 % difference between the average concentrations of three monitoring stations ($48\,\mu g/m^3$) and the mobile measurements around the three stations ($93\,\mu g/m^3$), both averaged for the measurement campaign period, 27 % can be explained by the difference in inlet height and distance to the street, and the remaining 21 % is due to the fact, that the monitoring stations are positioned away from pollution hot spots at crossroads according to WHO guidelines.

3.3 ~~Comparison between ground measurement and converted OMI observation~~

~~Satellite measurements are strongly affected by clouds, as clouds shield ground level NO_2. Hence, to compare with the LP DOAS data, OMI data with cloud fractions larger than 50 %, which were significantly influenced by clouds, were filtered out. LP DOAS data from 12:00 - 15:00 UTC, same as the OMI overpass time for Munich, were used for comparing with the average OMI data sets within 10 km and 50 km from the measurement site (). To reduce the impact of clouds and local spatial variations, we use monthly average data to compare. The uncertainty of the LP DOAS measurements, which was smaller than 1 ppb for a single measurement, is too small to be shown for monthly averages so the standard deviation within each month is shown in . Observation of LP DOAS and OMI both showed a similar annual trends, with higher NO_2 levels in winter and lower NO_2 levels in summer.~~

~~Comparing monthly means of LP DOAS overpass time measurements with OMI retrieved ground mixing ratios of NO_2 and VCDs within 10 km show correlation coefficients of 0.85 and 0.72, respectively (). OMI and the corresponding overpass time measurements of LP DOAS correlate well, indicating OMI measured reliable tendency of ground level NO_2. The discrepancy of correlation coefficients were mainly caused by the vertical profile used for the OMI retrieval and the conversion of VCDs to ground level mixing ratios. In order to show the influence of temporal averaging and assess the temporal representativeness of the governmental monitoring station data, monthly averages for all LP DOAS data and the data of three governmental monitoring stations are shown in as well. In addition, the data of the governmental monitoring stations were 15.4 ppb on average higher than the LP DOAS measurements. The Pearson correlation between the two data sets was 0.32. The low correlation may be due to the different measurement areas, measurement heights and measurement resolution.~~

~~Average variability of the NO_2 mixing ratios in winter (November to February) and summer (June to August) were 16.5 ppb and 5.5 ppb. The errors bars of OMI data do not overlap with the LP DOAS data for most months, neither within 10 km nor within 50 km, which demonstrated that there might be systematic errors in the conversion of VCDs to ground level mixing ratios or in the OMI retrieval process itself *Wenig et* al., 2008. As the previous study also suggests, lower OMI values over cities compared to ground measurements could be due to the OMI a-priori profile used for both, the VCD retrieval and converting the tropospheric NO_2 VCDs to ground level mixing ratios, was taking an average over a larger area, not only urban areas, but also~~

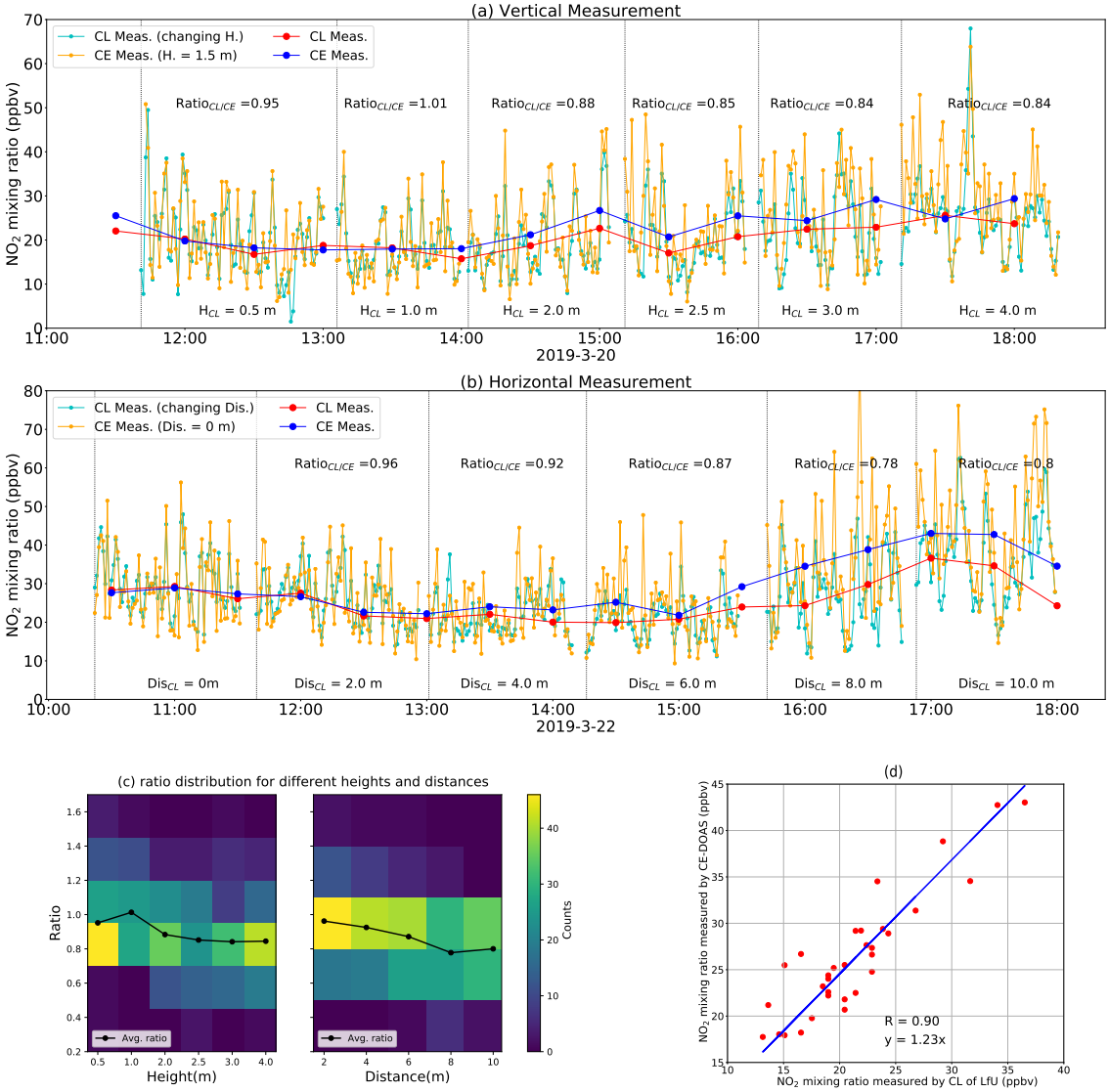

Figure 5. NO_2 measurements at Karlsplatz (Stachus), Munich, with different heights above ground (a) and distances to the main street (b). The ratio between 1-minute averaged CE DOAS and coinciding CL measurements were calculated individually for the different heights and distances. Half-hour averaged CE DOAS measurements (blue curve) were compared with the corresponding CL measurements (red curve). Averaged ratios for different heights and distances (as defined in (a)) are shown in (c), CE DOAS measurements at $1.5\,\mathrm{m}$ were averaged to $30\,\mathrm{min}$ intervals and compared with the half-hourly data of the governmental monitoring station at $4\,\mathrm{m}$ shown in (d)

~~rural areas with a lower ground level mixing ratio to total column ratios. A total underestimation for the ground level NO_2 of about 69 % can be observed. A similar result was found by *Kuhlmann et* al. [2015]. However, because of the good correlation it is safe to assume that relative temporal changes captured by the LP DOAS can be regarded as representative for area covered by OMI that spans the entire city of Munich.~~

4 Summary and conclusions

Mobile road measurements using CE DOAS and CAPS instruments combined with an algorithm for correcting the diurnal cycle were used in order to generate a consistent pollution map of the street level NO_2 concentration in Munich. This map is not only used to identify pollution hot spots but also to figure out how representative the existing NO_2 point measurements are for the whole city. Elevated NO_2 levels can be observed mostly on motorways and busy city roads, due to the emission of heavy duty vehicles or heavy traffic volume. When averaging the mobile measurements around the monitoring stations, we derived an average NO_2 concentration of $93\,\mu g/m^3$, whereas the three monitoring stations at the city center reported $48\,\mu g/m^3$ on average for the same time, so $48\,\%$ lower values. Our analysis shows that the different measurement height can account for $16\,\%$ difference (factor 0.84), and the distance of the sample inlets to the center of the street, where the mobile measurements took place explains the $13\,\%$ (factor 0.87) lower values. Accounting for these factors still leaves about $21\,\%$ that can be attributed to pollution hot spots like busy cross roads or tunnel exits. These hot spots are not covered by monitoring stations which is intentionally done in order to make the long term data less dependent on local events. Nevertheless, the differences observed in the presented study shows that point measurements are likely not representative for the NO_2 concentration in the whole city. Most network measurement sites are not capturing the concentrations people are exposed to when walking or driving at street level but are instead focusing on long term trends. Our study illustrates the importance of combining different measurement techniques to capture spatial and temporal patterns within a city and derive concentration values that are representative for the air most people breathe in.

The pollution maps generated in this project provide valuable information for future urban planning and the design of control measures of environment pollution. Furthermore, it can provide guidelines for identifying representative locations for air pollution monitoring stations in a city. Additionally, the observed spatial distribution of NO_2 concentrations are also beneficial to the validation of chemical transport models and assessment studies of the impact of air pollution on human health.

Author contributions. YZ, MW and JC designed the experiments. YZ, GK and XB carried them out. KLC simulate the results from GEOS-Chem model. YZ prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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