

Answers to the Referee 2

We thank the anonymous Referee 2 for positive review of the manuscript. The Referee 2 especially recommended to rewrite and shorten the Introduction and a part of Methodology section. To address the raised issues, we will answer the comments one by one and provide the changes made in the text. Due to major revision of the manuscript proposed by Referee 1, we will implement the changes to the new version of the manuscript.

- 1. The introduction section is unusually long with full of irrelevant information. I would suggest the authors modify the introduction section and re-write. Instead of describing the general impacts and roles of BC in the atmosphere (which are widely available in the literature), focus your description on the existing emission estimates, their problems, and the need for the methods which have been described in the manuscript. Two pages would be more than sufficient for the introduction.**

Thank you for this comment. We agree that by rewriting the Introduction section we can draw reader's attention to the importance of the presented method. In this regard, we have shortened the:

- introduction to black carbon (BC)
- information on worldwide BC emission inventories
- information about the existing approaches for mixing layer height (MLH) determination
- introduction to radon characteristics

Based on the comments of Referee 1, the information of modelled values of radon exhalation rate were included in the Introduction section (Pages 2 – 5).

1.Introduction

Black carbon (BC) ~~is~~ an important component of fine particulate matter in the atmosphere ~~and its most strongly light absorbing fraction. It is produced by incomplete combustion of various carbonaceous fuels, mainly fossil fuel and biomass. Due to its strong absorption of shortwave solar radiation, subsequent heating of the atmosphere and rapid adjustment effects on clouds and snow, it significantly contributes to the climate forcing by aerosols (Pöschl, 2005; Bond et al., 2013; IPCC, 2013) and is. Variety of other aerosols and precursor gases are emitted together with BC, which, after the internal mixing and aging in the atmosphere, alter BC optical properties (e.g. Cui et al., 2016; Pokhrel et al., 2017) and its atmospheric removal rate due to changes of hygroscopic properties of BC-containing particles (e.g. Zhang et al., 2015). In addition, BC is an important air pollutant, decreasing local air quality and is associated with undesirable health outcomes (Janssen et al., 2011; WHO, 2012).~~

Since BC is a chemically inert primary pollutant, it ~~is~~ can be used as a good measured indicator of emissions and can provide valuable information to authorities in the implementation and evaluation of air quality action plans, by indicating the strength of different emissions sources (e.g. Reche et al., 2011; Titos et al., 2015). On the other hand, emission inventories provide an important information for climate models by providing data about the changing pattern of BC emissions, its major sources and historical evolution. From the perspective of short-term local air quality prediction, improving local diurnal and seasonal pattern of BC emissions would greatly benefit the model prediction performance. ~~Although A~~atmospheric chemical transport models based on the fundamental description of atmospheric physical processes can improve the knowledge about temporal evolution of BC emissions at the modelled area, ~~but they~~ require comprehensive input data of atmospheric processes (Seinfeld and Pandis, 2016).

Bottom-up emission inventories rely on the information on the amount of used fuel combined with fuel-specific emission factors (e.g. Bond et al., 2007; Bond et al., 2013; Klimont et al., 2017). Although current emission inventories agree quite well on the main emission sources and regions, there exist significant uncertainties in the emission factors and activity data, used for emission calculation, with recent observationally constrained estimations much higher than the ones traditionally used (Sun et al., 2019). In contrast to bottom-up emission inventories, top-down constrained methods (such as inverse modelling) focus at minimising the difference between simulated pollutant concentration, based on estimated emission flux, and measured pollutant concentration (Brioude et al., 2013; Wang et al., 2016b; Guerrette and Henze, 2017). These methods can provide spatially and temporally better resolved assessment of pollutant emissions, including

BC, but are influenced by different sources of uncertainty, mainly from the insufficient evaluation of long-range transport of polluted air-masses.

~~Based on bottom up constructed emission inventories, BC emissions from energy related combustion have been increasing gradually from the beginning of industrial era from about 1 Tg in 1850 to 4.4 Tg in 2000 (Bond et al., 2007) and were dominated by contributions from different fuel types during technological evolution. Contribution from coal dominated until 1975, followed by biofuel and diesel fuelled engines more recently. Considering also open biomass burning, which accounts for 40 % of global BC emission, the total emissions in 2000 are estimated at about 7.5 Tg (Bond et al., 2013). According to the study of Klimont et al. (2017), where additional emission sources were considered, global BC anthropogenic emissions were estimated at about 6.6 and 7.2 Tg in 2000 and 2010, respectively, with a contribution of 0.7 Tg from Europe and Russia in 2010. Reduction of BC emission factors by implementation of cleaner technology partly offset the rapidly increasing energy consumption since 1950, resulting in slower growth of particulate matter emissions in comparison to CO₂. Although current emission inventories agree quite well on the main emission sources and regions, there exist significant uncertainties in the emission factors and activity data, used for emission calculation, with recent observationally constrained estimations much higher than the ones traditionally used (Sun et al., 2019). In contrast to bottom up emission inventories, top-down constrained methods (such as inverse modelling) focus at minimising the difference between simulated pollutant concentration, based on estimated emission flux, and measured pollutant concentration (Brioude et al., 2013; Wang et al., 2016b; Guerrette and Henze, 2017). These methods can provide spatially and temporally better resolved assessment of pollutant emissions, including BC, but are influenced by different sources of uncertainty, mainly from the insufficient evaluation of long range transport of polluted air masses.~~

According to the European Union emission inventory report (LRTAP, 2018), 0.2 Tg of BC were emitted in 2016 in the EU-28 region, with the dominant energy-related emissions from on-road and non-road diesel engines accounting for about 70 % of all anthropogenic BC emissions (Bond et al., 2013). Recently updated United States black carbon emission inventory (Sun et al., 2019) pointed out decreasing trend of BC emissions from 1960 to 2000, dominated by vehicle, industrial and residential sectors. Traffic related BC emission is dominating primary particulate matter (PM) emission especially in major cities (e.g. Pakkanen et al., 2000; Klimont et al., 2017). Recently, biomass combustion for residential heating has been promoted under the label of renewable fuel and additionally increased due to economic crises and increase of other fuel prices (Crilley et al., 2015; Denier van der Gon et al., 2015; Hovorka et al., 2015; Athanasopoulou et al., 2017). Although several studies report significant role of wood burning emissions on BC concentrations in Alpine valleys (Sandradewi et al., 2008b; Favez et al., 2010; Herich et al., 2014) and Scandinavian rural areas (Ricard et al., 2002; Aurela et al., 2011), increase in contribution of wood smoke to fine PM was noticed also in several large urban areas (Favez et al., 2009; Crippa et al., 2013; Fuller et al., 2014; Denier van der Gon et al., 2015; Hovorka et al., 2015; Helin et al., 2018; Zhang et al., 2019). Notable contribution of wood smoke was observed also in Slovenian urban (Ogrin et al., 2016) and rural areas (Wang et al., 2019), responsible for air quality deterioration especially in geographically constrained areas such as valleys and basins.

To assess the efficiency of abatement measures aiming to improve air quality, concentration of pollutants is usually measured before and after the measures are implemented, in order to quantitatively determine the reduction of pollutant concentration. However, this approach can be biased due to changes of micrometeorology of planetary boundary layer (PBL) which Besides the intensity of pollutant emission, micrometeorology of the planetary boundary layer (PBL) plays an important role in controlling time evolution of pollutant concentration. Therefore, assessment of BC emission rate requires decoupling of meteorologically driven variation from the dynamics of the sources. On diurnal timescales, atmospheric stability/dynamics plays a key role on the variability of primary inert pollutants (e.g. Quan et al., 2013; McGrath-Spangler et al., 2015; Tang et al., 2016), such as BC, and is affected by them (e.g. Ferrero et al., 2014). The evolution of the planetary boundary layer (PBL), is the lowest part of the troposphere, which is directly affected by the presence of the Earth's surface. Soon after sunrise is driven, convectively driven by convective heat transfer from the ground surface and by mechanical mixing (due to wind shear and surface roughness), which are is responsible for the formation of the turbulent mixing layer (ML), ML which grows by entraining the air from above and reaches its maximum depth in the late afternoon. Thus, the mixing layer height (MLH) strongly depends on the intensity of solar radiation reaching the ground. Entrainment of air from above takes place in a stable layer at the top of the ML, the so-called entrainment zone. The residual layer is formed after the decay of turbulence shortly before sunset, with its bottom portion transformed into a stable nocturnal boundary layer (SNBL) during

the night. SNBL is characterized by stable stratification with low mixing. The bottom 10 % of the boundary layer, either ML or SNBL, represents the surface layer, where turbulent fluxes and stress vary by less than 10 % of their magnitude (Stull, 1988). Anthropogenic air pollutants are generally emitted from the surface and trapped within the PBL, where their concentrations are controlled by the turbulent mixing (e.g. Quan et al., 2013; McGrath Spangler et al., 2015; Tang et al., 2016). Therefore, on diurnal timescales, atmospheric stability/dynamics (tendency of the atmosphere to resist or enhance the initial displacement of air parcels) plays a key role on the variability of primary inert pollutants, such as BC, and is affected by them (e.g. Ferrero et al., 2014).

Different approaches exist for MLH determination are reflected in different definitions found in literature (Seibert et al., 2000). One of generally adopted definitions relies on the vertical extent of dispersion of pollutants, which are released from the surface within 1 hour time period. However, since slope winds may influence vertical pollutant transport, this definition may be ambiguous at complex terrains such as Alpine valleys (Leukauf et al., 2016). MLH can be determined by indirect methods based on remote sensing using lidars (Caicedo et al., 2016; de Bruine et al., 2017) or radiosounding measurements, by applying different measures, such as Richardson number, Monin-Obukhov length or turbulence kinetic energy (Stull, 1988). These methods have usually low spatial and/or temporal resolution. Furthermore, interpretation of mixing in very stable conditions is challenging due to phenomena such as nocturnal jets and local flow perturbations (Williams et al., 2013), leading to non-reliable representation of stable atmospheric conditions by numerical weather or dispersion models.

Assessment of the BC emission rate requires decoupling of meteorologically driven variation from the dynamics of the sources. An alternative way to overcome the difficulty associated with the proper physical interpretation of micrometeorological properties of the ML and dispersion characteristics is the use of a tracer method. Naturally occurring noble radioactive gas radon (^{222}Rn) has been applied in the past for different studies. Radon characteristics, its emanation from rocks and its transport in rocks, soil (e.g. Etiope and Martinelli, 2002) and atmosphere (e.g. Griffiths et al., 2011; Williams et al., 2013) were comprehensively studied in the past. Radon was used to study: the study of long-range transport of air masses (Hansen et al., 1990; Crawford et al., 2007), and recently for studying PBL characteristics (e.g. Griffiths et al., 2013; Williams et al., 2013; Pal et al., 2015; Salzano et al., 2016; Vecchi et al., 2018), microclimate spatial variability (Chambers et al., 2016; Podstawczyńska, 2016) and impact assessment of atmospheric stability on local air pollution (Perrino et al., 2001; Chambers et al., 2015a; Chambers et al., 2015b; Crawford et al., 2016; Wang et al., 2016a; Williams et al., 2016). Good correlation, at least for the periods of thermally driven PBL convection, was observed in previous studies comparing effective MLH derived by the box model and MLH obtained by modelling approaches based on turbulence variables (Allegrini et al., 1994; Vecchi et al., 2018; Kikaj et al., 2019) or remote sensing techniques (sodar, lidar) (e.g. Salzano et al., 2016). Kikaj et al. (2019) successfully identified persistent inversion events in the Ljubljana basin based on ^{222}Rn measurements. The emission pattern of gaseous traffic related air pollutants in Bern (Switzerland), estimated by a box-model based on radon tracer, with an included advection term, showed an excellent correlation with traffic density (Williams et al., 2016). These studies imply that radioactive tracer method gives reliable information on the effective mixing layer height and indication of atmospheric stability (e.g. Perrino et al., 2001), which can be easily implemented in the environmental monitoring networks.

Radon is the only gaseous element in the ^{238}U radioactive decay chain, with its only significant sources in the natural environment being rocks and soil. After emanation from rock surfaces and soil grains, it is transported by diffusion and advection to the surface by carrier gasses (Etiope and Martinelli, 2002). Once exhaled from the Earth's surface, it is subjected to the mixing within the ML, thus experiencing the same extent of dispersion as other air pollutants emitted from ground-based sources. As a noble gas with a relatively short half-life (3.82 days), it represents an ideal tracer for the study of PBL processes. The strength of radon flux from the surface to the atmosphere, the so-called radon exhalation rate (E_{Rn}), depends mainly on the surface permeability and the radon potential (Karstens et al., 2015), which are controlled by geological and climatic characteristics of the area (e.g. Vaupotič et al., 2007; Vaupotič et al., 2010; Kardos et al., 2015; Karstens et al., 2015). Surface permeability is controlled by geological setting—lithological (rock type) (e.g. Kardos et al., 2015), structural characteristics (presence of fault zones) (e.g. Vaupotič et al., 2010) and soil moisture. Fault zones can act as a pathway for different gaseous species especially in the geologically active environments, leading to anomalies in E_{Rn} , whereas E_{Rn} above karst caves and fractured rocks can experience high spatial and temporal variability (Vaupotič et al., 2010; Gregorič et al., 2014). Since radon is poorly soluble in water, its only sink is the radioactive decay, which can

~~be neglected on hourly bases.~~ As reported by Vaupotič et al. (2007) measured E_{Rn} on Slovenian territory spans over large range from few tens to several hundred $mBq\ m^{-2}\ s^{-1}$. [Karstens et al. \(2015\) reported modelled radon exhalation rate on European scale, which varies in the range from 20 – 40 \$mBq\ m^{-2}\ s^{-1}\$ and 45 – 50 \$mBq\ m^{-2}\ s^{-1}\$ in the south-western part of Slovenia in winter and summer, respectively. The range of \$E_{Rn}\$ in the central part of Slovenia is 10 – 25 \$mBq\ m^{-2}\ s^{-1}\$ and 25 – 35 \$Bq\ m^{-2}\ h^{-1}\$ for winter and summer, respectively.](#) Radon exhalation rate is usually considered constant on short temporal scales in areas with homogeneous geologic characteristics (Pearson and Jones, 1965). However, exhalation of Rn is a complex process which can be assessed with different modelling approaches. Salzano et al. (2016) showed, that the error in the modelled effective MLH by considering constant radon source can be up to 10 %. Local heterogeneity of E_{Rn} due to heterogeneous soil permeability (within few meters range) is homogenised in the thin atmospheric layer (~0.5 m) close to the ground and does not represent a significant concern for measurements above this height. [Due to continuous source from the surface, radon concentration profiles in the SNBL can exhibit strong gradients, resulting in higher radon concentration when measurements are conducted closer to the ground, especially in the SNBL conditions.](#) → On a seasonal scale, however, E_{Rn} decreases with the presence of snow cover, frozen soil or during and shortly after (on time scale of few days) rainy periods, due to reduced surface permeability, thus representing one of the main sources of uncertainty in the box model. It is also worth noting, that reliable exhalation measurements (used for the box model) should be conducted in a broad network in the extent of modelled area in different periods of year, so the seasonal changes in soil permeability would also be considered. The seasonal changes of E_{Rn} were already pointed out in different studies (Karstens et al., 2015; Salzano et al., 2016; Williams et al., 2016; Chambers et al., 2019).

The aim of this paper is to determine the BC emission rate apportioned to traffic and biomass burning sources, its diurnal pattern and monthly variation for two distinct locations in Slovenia (Europe), which differ from the point of view of their natural characteristics (geology, geomorphology, meteorology) and urban environment (urban and rural background). Both sites are subjected to their own pattern of air pollution episodes which will be addressed and interpreted based on Eulerian box model. The effective mixing layer height will be reproduced for both sites based on Rn measurements, taking into account seasonally resolved E_{Rn} , then used for decoupling meteorologically driven changes of measured BC concentration from the one resulting from the source dynamics. The highly time-resolved and source apportioned BC emission rate (E_{BC}) represents an essential information for short-term forecasts of air pollution episodes, as well as for the evaluation of the efficiency of air quality abatement measures and their potential adaptation. Temporal variation of BC concentration will be highlighted from the point of view of PBL evolution. A list of acronyms and symbols used in this paper is given in Table 1.

2. **Section 2.1 is also full of unnecessary information. Page 6, line 13-31: please reduce the content. There is no need to describe the population, growth, and implementation of various plans by the municipality. Please merge the ‘measurement locations’ and ‘geological setting’ together.**

As proposed by the Referee 2, the paragraph “Measurement locations” and “Geological setting” were merged and the section was shortened. However, from the point of view of interpretation of obtained results, we think it is important to provide an information regarding population density and characteristics of BC sources for both locations. When implementing abatement measures, such as traffic restrictions, the emission rate is the quantity which quantitatively demonstrates the efficiency of the measures taken.

The section “2.1 Measurement locations” was changed as follows:

2.1 Measurement locations

Two distinct measurement locations were selected for this study. The first one is located in the urban area of Ljubljana (LJ, capital city of Slovenia), which lies in the central part of Slovenia (Europe) (Figure 1), situated in a sub-Alpine basin. The second measurement location was in a small town Ajdovščina (AJ), located in the Vipava valley in the western part of Slovenia. Measurement campaigns lasted from autumn 2016 to spring 2017 (November 23, 2016 to May 20, 2017) in Vipava valley and from winter 2017 to summer 2017 (February 1, 2017 to June 8, 2017) in Ljubljana. Due to its basin location, Ljubljana is characterised by poor ventilation and frequent occurrence of persistent temperature inversions (Kikaj et al., 2019), which constrains pollutants emitted from surface sources within the limited air volume inside the basin. During stable atmospheric conditions, especially in the SNBL, a thin layer of drainage winds (colder air, adjacent to the ground, flowing downhill under the influence of gravity) and a flow of air from the edge of the city towards the centre (formed due to the heat island) governs air circulation within the Ljubljana basin (Stull, 1988; Ogrin et al., 2016). ~~During the measurement campaign, around 10 cm of snow cover was present from January 14 to February 2, 2017.~~ Ljubljana is characterised by temperate continental climate, with a significant seasonal temperature cycle.

The population of the wider Ljubljana basin is around 500,000, of which 287,000 live in the urban municipality of Ljubljana. Much more than 100,000 daily commuters from other municipalities represent additional traffic flow on working days: (Ogrin et al., 2016). ~~Rapid growth of automobile use is observed in the last few decades, leading to daily traffic jams inside the city and its surroundings. In the recent years, the local municipality has implemented different measures of sustainable mobility in order to improve the air quality. In particular, the traffic restriction in the major road in the city centre has led to 70% decrease of local BC concentrations (Titos et al., 2015).~~ Besides traffic-related air pollution, emissions from combustion of biomass fuel for residential heating are a significant source of particulate concentrations in the whole country (Gjerek et al., 2018), not only in rural areas, but in Ljubljana as well. Although district heating is provided in several areas of the city, the use of wood boilers and fireplaces is a common practice.

The population of the second area of interest, including surrounding villages is relatively small, with ~~about 19,000 residents living in the municipality of Ajdovščina, of which~~ about 7,000 residents ~~live~~ living in the town of Ajdovščina. The Vipava valley is confined to the north by the steep ridge, which rises up to about 1000 m a.s.l., and to the south by the Karst plateau with an average altitude of about 300 m. Due to the complex topography, the valley usually experiences two extreme cases of atmospheric stability conditions. On one side, stable atmospheric conditions can last for several days, leading to the formation of strong vertical aerosol gradients, which are followed by frequent occurrences of strong downslope Bora wind (Mole et al., 2017; Wang et al., 2019). A highway connecting the central part of Slovenia with Italy runs through the valley on the southern border of the town, around 800 m away from our measurement site. The Mediterranean climate of this area is responsible for mild winters and warm summer season, with residential heating mainly limited to the cold season, from November to February, with biomass fuel being the primary source of energy.

Geological setting

From geological point of view, the city of Ljubljana lies in the neotectonic basin with extensive and thick accumulations of Quaternary glaciofluvial sediments on the northern and central parts (gravel and conglomerate), whereas the south-western part of Ljubljana basin is filled with lacustrine and paludal sediments (Janža et al., 2017). The maximum thickness of sediments is around 170 m. Non-

consolidated Quaternary sediments are permeable enough to allow spatially and temporally homogeneous Rn exhalation rate.

The geological structure of the broader area of Vipava valley results from Tertiary thrust of Cretaceous limestone, which forms the steep north-eastern ridge of the valley, on the Eocene flysch rocks, forming the valley floor. Flysch rocks consist of alternating layers of marlstone and carbonatic sandstone. Due to physical weathering of the limestone, a large amount of limestone scree material has been formed and deposited on the underlying flysch rocks on the slopes of the north-eastern ridge. Valley floor is covered by clayey weathered residual of flysch rocks with fine flysch scree (Jež, 2007). Spatially homogeneous Rn exhalation rates can be expected along the valley.

3. Page 8, line 3-30, already available in the literature, not needed specifically. Just cite the literature and remove the theoretical information.

We agree to remove the theoretical information about BC source apportionment from the main text, since this allows to keep the focus of the paper on the application of radon box model and results of BC emission rates (E_{BC}). However, the parameters used for source apportionment may significantly influence the results of E_{BC} . Therefore we decided to shorten the text and move it to the Supplement.

The section “2.2 Black carbon measurements and source apportionment” on Page 8, lines 3 – 30 was corrected.

Aethalometer measurements at different wavelengths provide an insight in the chemical composition of light absorbing particles. The so called Aethalometer model (Sandradewi et al., 2008a) was used to apportion BC to traffic (BC_{TR}) -and biomass burning (BC_{BB}) sources. The model uses an a priori assumed pair of absorption Ångström exponents (AAE) for traffic (AAE_{TR}) and biomass burning (AAE_{BB}) to determine the contribution of both sources, ~~which were set to 1.0 and 2.0 for AAE_{TR} and AAE_{BB}, respectively. A narrow range of AAE_{TR} (0.8 – 1.1) values is reported in the literature, whereas larger AAE_{BB} values (from about 1.5 up to 3.5) in the wider range are characteristic for biomass burning sources (Kirchstetter, 2004; Saleh et al., 2013; Garg et al., 2016; Zotter et al., 2017). Higher values of AAE_{BB} result from enhanced light absorption in the near-UV and blue part of the spectrum caused by organic carbon species, present in biomass smoke. Source specific AAE can be independently determined using auxiliary measurements of OC/EC, and ¹⁴C (Sandradewi et al., 2008a; Zotter et al., 2017), or biomass burning tracers like levoglucosan (Favez et al., 2010; Herich et al., 2014; Hellén et al., 2017; Helin et al., 2018). Since independent measurements allowing the determination of the AAE pair representative for our measurement locations were not available, the most suitable AAE pair was estimated according to the commonly used AAE values published in the literature, by considering overall distribution of AAE (Figure S1) for each measurement location and the corresponding diurnal variation of traffic (BC_{TR}) and biomass burning related BC (BC_{BB}) (Figure S2). AAE was calculated using the Eq. 1 for 470 nm and 950 nm wavelengths, where b_{abs} stands for the absorption coefficient at 470 nm and 950 nm.~~

$$AAE = \frac{\ln\left(\frac{b_{abs(470)}}{b_{abs(950)}}\right)}{\ln(950/470)} \quad (1)$$

~~By taking into account equations provided by Sandradewi et al. (2008a), the BC_{BB} and BC_{TR} were finally calculated using the Eq. 2 and 3, respectively.~~

$$BC_{BB} = \frac{\frac{b_{abs(470)}}{b_{abs(950)}} - \left(\frac{950}{470}\right)^{AAE_{TR}}}{\left(\frac{950}{470}\right)^{AAE_{BB}} - \left(\frac{950}{470}\right)^{AAE_{TR}}} \times BC \quad (2)$$

$$BC_{TR} = BC - BC_{BB} \quad (3)$$

~~AAE_{TR} / AAE_{BB} pair of 1.0 and 2.0, respectively, was chosen for both measurement locations. Further discussion on the choice of AAE pair used for source apportionment is provided in the Section 1 of the Supplement.~~

4. Page 9, line 12: Please add the full form of FFT in the list of abbreviations in Table 1.

Table 1 was updated with adding the FFT to the list of abbreviations.

Acronym/Symbol	Definition	Units
λ'_s	temporal decay constant	h^{-1}
AAE	absorption Ångström exponent	
AAE _{BB}	biomass burning related AAE	
AAE _{TR}	traffic related AAE	
AJ	Ajdovščina location	
ARSO	Slovenian Environmental Agency	
b_{abs}	absorption coefficient	Mm^{-1}
BC	black carbon concentration	ng m^{-3}
BC _{BB}	biomass burning related black carbon concentration	ng m^{-3}
BC _{TR}	traffic related black carbon concentration	ng m^{-3}
C	multiple scattering parameter	
C _{Rn}	radon activity concentration	Bq m^{-3}
C _s	species concentration	
E _{BC}	black carbon emission rate	$\mu\text{g m}^{-2} \text{h}^{-1}$
E _{BB}	black carbon emission rate from biomass burning sources	$\mu\text{g m}^{-2} \text{h}^{-1}$
E _{TR}	black carbon emission rate from traffic sources	$\mu\text{g m}^{-2} \text{h}^{-1}$
EMEP	The European Monitoring and Evaluation Programme	
E _{Rn}	radon exhalation rate	$\text{Bq m}^{-2} \text{h}^{-1}$
E _s	species emission rate	
FFT	Fast Fourier transform	
GDAS	Global Data Assimilation System	
GOL	Golovec Astronomical and Geophysical Observatory	
h	effective mixing layer height	m
LJ	Ljubljana location	
ML	mixing layer	
MLH	mixing layer height	m
NOAA-ARL	NOAA Air Resources Laboratory	
OT	Otlica Meteorological observatory	
PBL	planetary boundary layer	
PM	particulate matter	
SNBL	stable nocturnal boundary layer	
T	air temperature	°C
wd	wind direction	
ws	wind speed	m s^{-1}
γ_s	spatial decay constant	m^{-1}
σ_{air}	mass absorption cross section	$\text{m}^2 \text{g}^{-1}$

5. Section 2.4: It is recommended to provide a scatterplot of modelled-MLH with GDAS also in supplementary file.

Since the same comment was also given by Referee 1, we have improved the description of methodology used for MLH comparison and the results, including the scatterplot, were added to the Supplement. A more detailed reply is provided in the Answers to Referee 1.

6. **Page 14, line 23: Authors mention that the Average Radon activity concentration was similar at both measurement locations, i.e., $15 \pm 11 \text{ Bq m}^{-3}$ and $14 \pm 10 \text{ Bq m}^{-3}$. At the same time, the authors also mentioned that it was slightly lower in the spring $13 \pm 9 \text{ Bq m}^{-3}$ and $12 \pm 8 \text{ Bq m}^{-3}$. Consider the standard deviations in the data, I do not see any difference in the data. Authors should check whether these differences are statistically significant or not and add a line on it.**

We agree with the observation pointed out by the Referee 2. There is in fact no significant difference in the seasonal averages of radon concentration measured at both locations. The text was corrected accordingly.

Page 14, lines 23 to 28:

Average radon activity concentration derived from hourly measurements (Figure 3) was similar at both measurement locations, $15 \pm 11 \text{ Bq m}^{-3}$ and $14 \pm 10 \text{ Bq m}^{-3}$, ~~measured in winter~~ and $13 \pm 9 \text{ Bq m}^{-3}$ and $12 \pm 8 \text{ Bq m}^{-3}$ in spring, in Ljubljana and Vipava valley, respectively. ~~Slightly lower average C_{Rn} was characteristic for spring: $13 \pm 9 \text{ Bq m}^{-3}$ and $12 \pm 8 \text{ Bq m}^{-3}$, respectively for both locations~~ (Table 4). These values are above annual average outdoor radon concentration of 10 Bq m^{-3} reported by UNSCEAR (2000) for the continental areas. Due to limited atmospheric mixing, higher winter concentrations are usually observed. However, decrease of atmospheric C_{Rn} by more efficient atmospheric mixing is compensated by increased radon exhalation rate in the warmer season, which complies with the seasonal variation, which usually results in higher winter concentrations due to limited atmospheric mixing.

7. **Page 15, line 14: Despite significantly higher.....25% higher in LJ than in AJ. What is meant by only 25% higher? And how population is a factor here? It looks highly ambiguous statement. The authors should consider removing it.**

The statement was intended to point out the difference between BC concentration at both locations, based on the assumed stronger BC emissions in the area of denser population (Ljubljana) and subsequent traffic density. Since the average BC concentrations are discussed further on in the same section, this statement was removed.

8. **Figure 1 should be modified significantly. The background map items are almost invisible.**

Thank you for this observation. Fig. 1 was modified for clear visibility of all important items.

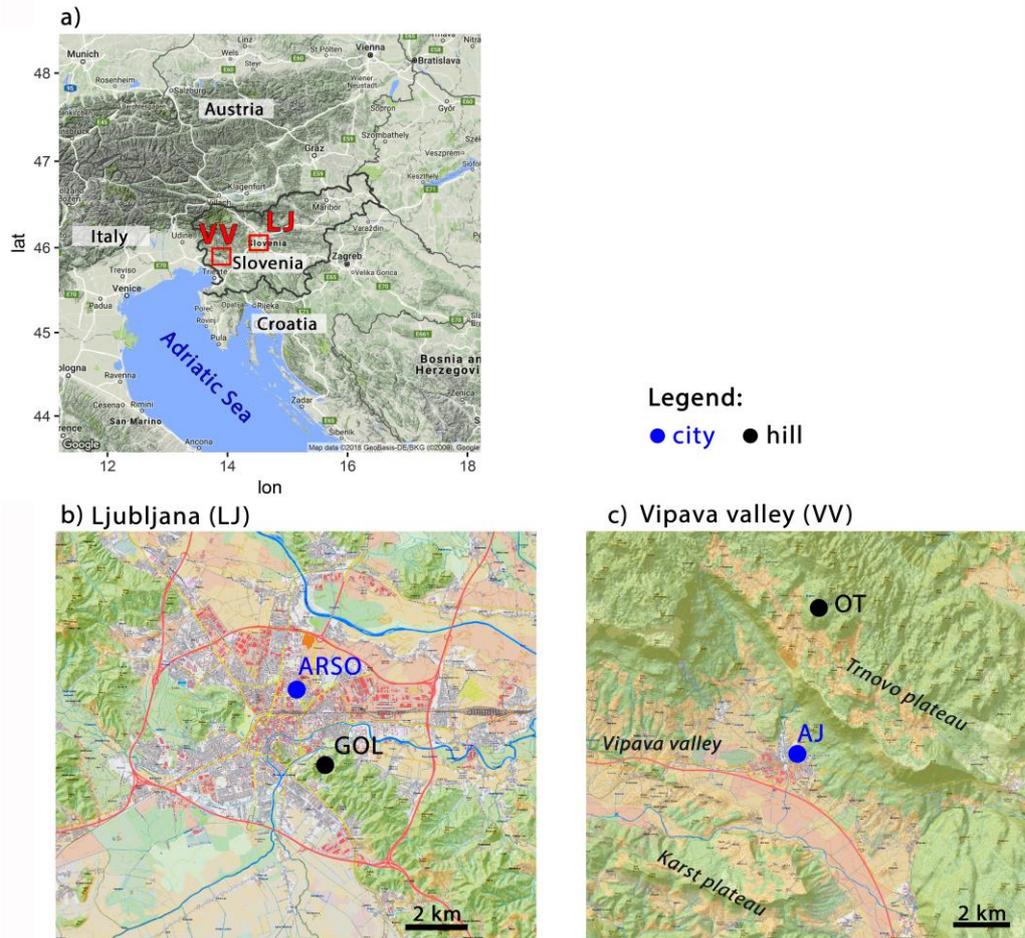


Figure 1: Map of Slovenia (a) with marked areas of measurement sites Ljubljana (LJ) and Vipava valley (VV). b) [The city area](#) of Ljubljana with urban background (ARSO) and hill (Golovec – GOL) measurement sites. c) Area of the Vipava valley with urban background (Ajdovščina – AJ) and hill (Otlica – OT) measurement sites (Source: Map data ©2018 GeoBasis-DE/BKG (©2009) Google [and OpenStreetMap](#))