Atmospheric Environment: X 2 (2019) 100021

Contents lists available at ScienceDirect

Atmospheric Environment: X

journal homepage: www.journals.elsevier.com/atmospheric-environment-x

Climate change impact on the dispersion of airborne emissions and the resulting separation distances to avoid odour annoyance

Martin Piringer[a,*], Werner Knauder[a], Ivonne Anders[b], Konrad Andre[b], Werner Zollitsch[c], Stefan J. Hörtenhuber[c], Johannes Baumgartner[d], Knut Niebuhr[d], Isabel Hennig-Pauka[e], Martin Schönhart[f], Günther Schauberger[g]

[a] *Department of Environmental Meteorology, Central Institute of Meteorology and Geodynamics, Vienna, Austria*
[b] *Department for Climate Research, Central Institute of Meteorology and Geodynamics, Vienna, Austria*
[c] *Austria Division of Livestock Sciences, Department of Sustainable Agricultural Systems, University of Natural Resources and Life Sciences, Vienna, Austria*
[d] *Institute of Animal Husbandry and Animal Welfare, University of Veterinary Medicine, Vienna, Austria*
[e] *University Clinics for Swine, Department for Farm Animals and Veterinary Public Health, University of Veterinary Medicine, Vienna, Austria*
[f] *Institute for Sustainable Economic Development, Department of Economics and Social Sciences, University of Natural Resources and Life Sciences, Vienna, Austria*
[g] *WG Environmental Health, Unit for Physiology and Biophysics, University of Veterinary Medicine, Vienna, Austria*

HIGHLIGHTS

- Climate change has an impact on dispersion parameters.
- Impact on dispersion parameters is site-dependent.
- A slight increase of the separation distances is to be expected in the future.

ARTICLE INFO

Keywords:
Stability class
Separation distance
Odour
Climate change
Livestock

ABSTRACT

In a changing climate it is expected that the predicted temperature rise will also affect wind conditions and parameters needed to determine atmospheric stability. In this paper we investigate how the changes in wind conditions and stability will affect the dispersion of airborne emissions. For odorous substances, the separation distances around livestock farms are calculated to protect the neighbourhood from odour annoyance. This is done for two Central European sites in Austria with different meteorological conditions, situated in areas with considerable livestock activity, namely around Wels in the north-alpine foreland and at Feldbach in the Raab valley south of the Alpine chain. Two climate scenarios have been used, namely the time period 1981–2010 (present climate) and the period 2036–2065 (future climate). Slight changes in wind and stability conditions for the two sites have been obtained, influencing the changes in separation distances. Although these are only minor, they show some interesting differences not expected a priori. We conclude that the climate change signal has mostly only a low impact on the separation distances. This will not substantially affect the zoning and licensing of livestock buildings in Austria in the course of this century.

1. Introduction

Beside many other areas, the impact of anthropogenic warming on air quality is a hot topic (Zlatev et al., 2011). In a more general perspective of air quality issues, this impact is described by the frequency of the air stagnation (Horton et al., 2014; Huang et al., 2017). Climate change can affect ambient air pollution in a number of ways through changes of the emission (e.g. increase of the emission rate by higher temperature and wind velocity shown for livestock buildings by Schauberger et al., 2014; Schauberger et al., 2018 and waste treatment plants by Schauberger et al., 2011)), of the dispersion in the atmosphere, in the atmospheric chemistry, and in the deposition (Athanassiadou et al., 2010). Interactions between anthropogenic warming and the emission, shown for fattening pigs by Schauberger

* Corresponding author.Central Institute of Meteorology and Geodynamics, Division Costumer Service/Department of Environmental Meteorology, Hohe Warte 38, A 1190, Vienna, Austria.
E-mail address: martin.piringer@zamg.ac.at (M. Piringer).

https://doi.org/10.1016/j.aeaoa.2019.100021
Received 4 July 2018; Received in revised form 4 February 2019; Accepted 8 February 2019
Available online 21 February 2019

et al. (2014), were faded out in this paper to underline the impact of climate change on the relevant dispersion parameters. To calculate the ambient concentration of airborne emissions, dispersion models are in use as the gold standard (Leelőssy et al., 2014). These models are directly driven by standard meteorological parameters like wind speed and direction, but also by the atmospheric stability, which can be derived by calculation schemes using several other parameters (radiation balance, cloud cover, time of the day, solar height, variability of the wind speed etc.). The influence of the climate change signal on these dispersion parameters will be analysed in this article.

In a second step the outcome of the dispersion calculation is analysed. We select the emission of odorous substances from confined livestock production, because the perception of odour is one of the most frequent causes for environmental complaints. The ambient odour concentration is evaluated by the separation distance, which is determined by percentiles between 85% and 98% of the exceedance probability of a preselected threshold (Brancher et al., 2017; Sommer-Quabach et al., 2014). The direction dependent separation distance between odour sources and residential areas is used to divide the circumjacent area around a source in a zone which is protected from annoyance and a zone closer than the separation distance where annoyance can be expected. This concept has the advantage that those meteorological conditions are considered most which represent higher pollution levels (higher ambient odour concentration) than the mean value (Hou and Wu, 2016).

For assessments of an expected future climate change only climate models can give an estimate in what way the observed trends might continue. For the current investigation, two climatic reference scenarios (for Upper Austria and South-eastern Styria) and future scenarios have been compiled. It is expected that the predicted temperature rise will also affect wind conditions and parameters needed to determine atmospheric stability. These parameters are used by dispersion models to predict ambient concentrations of trace substances, depending on the emission rate and the meteorological conditions. Of the latter, wind direction, wind speed, atmospheric stability and the mixing height are most relevant. Atmospheric stability is seen as the most critical parameter, as it can be determined in various ways, all with a considerable amount of uncertainty (e.g. Piringer and Schauberger (2013); Piringer & Joffre, Eds. (2005).

Based on the provided climate parameters, discrete stability classes will be calculated by two different methods (KTA 1508 (2006); VDI 3782 part 1 (2009)). All meteorological parameters which run the dispersion model are available on an hourly basis. A statistical analysis will reveal different sensitivities of these methods to climate change, depending on the meteorological parameters used. The changes between the current and the future scenarios are of particular interest.

The changes in meteorological input data between the reference years and the selected investigation areas will result in differences of separation distances to protect the neighbourhood from odour annoyance (Piringer et al. (2015); Schauberger et al. (2002)). With respect to the selected climate scenarios and the variety of the stability schemes, a bandwidth of affected areas and separation distances will result, with a possible judgement if and how, in the future, livestock husbandry will have to adapt to climate change, e.g. with impacts on today's licensing processes.

2. Materials and methods

2.1. Meteorological data

The two focus regions Upper Austria north of the Alpine chain (centred around Wels, 48.16 °N, 14.07 °E) and South-eastern Styria (centred around Feldbach, 46.95 °N, 15.88 °E) are located within class Cfb (warm temperature, fully humid, warm summers) following the climate classification of Köppen and Geiger (c.f. Kottek et al., 2006). Both are representative for large areas in Central Europe excluding the Alps. According to Chimani et al. (2016), for the area in Upper Austria, the annual mean temperature is 8.8 °C and the mean annual precipitation amount is 979 mm/year. Following the “business-as-usual”-climate change scenario the temperature might increase by $\sim +1.4$ °C ($\sim \pm 0.5$ °C) until the middle of the century. The number of hot days (daily maximum temperature $\geq$ 30 °C) is expected to increase in this region to between 4.7 and 5.0 days/year in the middle of the century compared to the reference period of 1971–2000 with a mean value of 3.3 hot days/year. The region in South-eastern Styria around Feldbach is, compared to the Upper Austria area, slightly more influenced by the Mediterranean and the Eastern European climate. In the recent time period 1981–2010 a mean temperature of 9.5 °C and an annual mean precipitation amount of 830 mm/year has been observed. In a “business-as-usual”-scenario a temperature increase of 1.5 °C might be possible. For this region the number of hot days is 6 days in the recent climate, but might increase by up to 8 days until the middle of this century.

For this study meteorological parameters are needed which are not observed and which are therefore derived from regional dynamical climate model output. With respect of observed temperature, a so-called reference year has been combined from model output based on a COSMO-CLM (Rockel et al., 2008) simulation for the time period 1981–2010 covering the Alpine Region in spatial resolution of 9 km forced by ERA-interim reanalysis data (Dee et al., 2011). Out of this simulation, the meteorological parameters wind direction, wind speed, radiation balance and total cloud cover needed for the current investigation (Sections 2.2 and 3.1) can be provided for the two different regions on an hourly basis for one year which is representative for a climate period of 1981–2010. The climate change signal has been derived from EURO-CORDEX regional climate model simulations only for the mean temperature, which is the most robust signal (Smiatek et al., 2016; Jacob et al., 2014). The ensemble mean temperature change was added to the observed temperature, and again out of the regional climate model simulation, a second reference year has been derived which is representative for the future climate period of 2036–2065. To use model output directly from EURO-CORDEX climate simulations for the future is not possible as the temporal resolution of these data is only daily. Furthermore, the meteorological parameters needed in this study are not available in the EURO-CORDEX ensemble (see also www.euro-cordex.net).

2.2. Dispersion model, stability classes, and source data

The dispersion model LASAT (Janicke Consulting, 2013) simulates the dispersion and the transport of a representative sample of tracer particles utilizing a random walk process (Lagrangian simulation). It computes the transport of passive trace substances in the lower atmosphere (up to heights of about 2000 m) on a local and regional scale (up to distances of about 150 km). LASAT is usually run with the Klug-Manier stability scheme (TA Luft, 2002). LASAT has been evaluated using test data sets for different applications (e.g. Hirtl et al., 2007; Hirtl and Baumann-Stanzer, 2007; Baumann-Stanzer et al., 2008; Piringer and Baumann-Stanzer, 2009; Schatzmann et al., 2010; Baumann-Stanzer et al., 2014)). More references concerning LASAT model evaluation can be found at www.janicke.de.

Based on the provided climate parameters, two methods to determine stability classes can be used. The first one is the German Klug/Manier (K-M) stability scheme (TA Luft, 2002). A detailed description of the determination of Klug/Manier stability classes is found in VDI 3782 part 1 (2009). Table 1 shows the basic scheme.

When there are only cirrus clouds present, the total cloud cover is reduced by 3/8. Cirrus clouds can be determined by observation of cloud type or the measured cloud base height. In addition to the correction of cirrus clouds, several further rules for season and time of day apply which are of concern also for the unstable class V which does not show up in the basic scheme displayed in Table 1.

Table 1
Basic scheme for the determination of Klug/Manier stability classes.

Wind speed υ_{10} at 10 m height (z_0 = 0.1 m) in ms^{-1}	Nighttime hours		Daytime hours		
	Total cloud cover (oktas)		Total cloud cover (oktas)		
	0/8 to 6/8	7/8 to 8/8	0/8 to 2/8	3/8 to 5/8	6/8 to 8/8
≤1.2	I	II	IV	IV	IV
1.3 to 2.3	I	II	IV	IV	III/2
2.4 to 3.3	II	III/1	IV	IV	III/2
3.4 to 4.3	III/1	III/1	IV	III/2	III/2
≥4.4	III/1	III/1	III/2	III/1	III/1

Klug/Manier classes are numbered from I to V and represent a simplified characterization of the turbulence situation:

- Dispersion categories V and IV comprise very unstable and unstable conditions, meaning good vertical mixing in the boundary layer. They do not occur during night-time. Category V occurs only between May and September.
- Dispersion categories III/2 and III/1 are classified as neutral. III/2 occurs predominantly at daytime, III/1 predominantly at night-time and during sunrise and sunset. These categories are typical for cloudy and windy conditions.
- Dispersion categories II and I comprise stable and very stable conditions, mostly, but not exclusively at night.

The other scheme used is described in KTA 1508 (2006). It uses again provided climate parameters, the radiation balance in combination with wind speed. The six Pasquill stability classes A, B, C, D, E and F are assigned to Klug/Manier classes because LASAT uses the Klug/Manier stability scheme as follows: A … V, B … IV, C … III/2, D … III/1, E … II, F … I. The stability class is then determined according to Table 2, using the radiation balance and the wind speed in 10 m.

For all model runs, the same source data are used (Table 3). The source is assumed non-buoyant, i.e. the effective stack height is equal to the physical stack height. The source is a livestock building, 3 m high, 120 m long and 30 m wide. Along the main axis of the building, 9 stacks are located, centred at the middle of the roof, with an equal distance of 13.3 m. Each stack has a volume flow rate of 20 000 m^3h^{-1} and an odour emission rate of 1500 $ou_E\ s^{-1}$. The total emission rate of 13 500 $ou_E\ s^{-1}$ corresponds to 1930 fattening pigs, 113 000 broilers and 940 cattle (VDI 3894 Part 1, 2011).

Table 2
Scheme to determine stability classes based on the radiation balance and the wind speed according to KTA 1508 (2006).

Wind speed υ_{10} at 10 m height	Radiation balance in Wm^{-2}				
	Limits of catagories				
in ms^{-1}	A/B	B/C	C/D	D/E	E/F
0 to 0.9	214	125	60	−2	−9
1.0 to 1.9	214	126	60	−4	−13
2.0 to 2.9	301	162	60	−6	−21
3.0 to 3.9	400	232	63	−12	−34
4.0 to 4.9	495	305	67	−28	−55
5.0 to 5.9	–	376	84	−55	–
6.0 to 6.9	–	450	108	–	–
7.0 to 7.9	–	–	150	–	–
8.0 to 9.9	–	–	240	–	–
≥10.0	all values catagory D				

Example: If the conditions 2.0 ms^{-1} ≤ u_{10} < 3.0 ms^{-1} and 162 Wm^{-2} ≥ radiation balance > 60 Wm^{-2} are fulfilled, then category C is used. Before applying the table, correct the values of u_{10} to one decimal.

Table 3
Source data for dispersion calculations; total emission (9 point sources).

Stack height	[m]	5.0
Stack diameter	[m]	1.88
Outlet air velocity	[ms^{-1}]	2.0
Volume flow rate	[m^3h^{-1}]	180 000
Temperature	[°C]	0
Odour emission rate	[$ou_E\ s^{-1}$]	13 500
Concentration	[$ou_E\ m^{-3}$]	270

2.3. Determination of separation distances

The separation distances are calculated on the basis of hourly values of the ambient odour concentration, calculated by the dispersion model. By the use of an odour impact criterion OIC, separation distances are defined for a certain protection level. We selected the German guide line (GOAA, 2008). The one year time series is evaluated by the exceedance probability of 10% for pure residential areas and 15% for weaker protected areas like commercial/industrial and agriculturally dominated areas, each in combination with a preselected odour concentration threshold of 1 $ou_E\ m^{-3}$. For the area closer than the direction-dependant separation distance, annoyance can be expected, for areas further away a far reaching odour protection can be expected.

When using a dispersion model like LASAT to calculate separation distances to protect the neighbourhood from odour annoyance, a transformation of the hourly mean values calculated by the model to short-term concentrations relevant for human odour perception is necessary. For Austria, to determine the short-term peak concentrations required for the assessment of odour perception, the authors developed a peak-to-mean approach depending on atmospheric stability (Schauberger et al. (2000) and Piringer et al. (2007); in Piringer et al. (2015), the latest version is described in detail). The resulting site-independent peak-to-mean attenuation curves are shown in Fig. 1.

The peak-to-mean ratios depend strongly on the stability class. For unstable conditions (Klug/Manier classes V and IV) the peak-to-mean factors, starting at rather high values near the source, rapidly approach 1 with increasing distance (Fig. 1). This is in agreement with the premise that vertical turbulent mixing can lead to short periods of local high ground-level concentrations, whereas the ambient mean concentrations are low. For neutral conditions (classes III/2 and III/1), the decrease of the peak-to-mean ratio is more gradual with increasing distance, because vertical mixing is reduced and horizontal diffusion is dominating the dispersion process. The peak-to-mean ratio in 100 m is then between 2 and 4. For stable conditions (classes II and I, identical curves due to identical values in Table 2 in Piringer et al., 2015), the peak-to-mean ratio exceeds 2 only near the source. Stability class III/2 ("neutral") gives the largest peak-to-mean factor for all distances between 100 and 500 m.

3. Results

This section is split in three parts. First, the climatic changes for those meteorological parameters which are needed to determine atmospheric stability classes are analysed. This is followed by the presentation of the changes in the frequency of stability classes in Section 3.2. In these first two Sections, the meteorological parameters are analysed which are needed to run a dispersion model. In Section 3.3, the outcome of the dispersion model, the changes of separation distances, are shown. A discussion of these findings follows in Section 4.

3.1. Changes in meteorological conditions

The two methods used to determine stability classes (Section 2.2) need information on wind direction and wind speed, so the change in these data is shown first. The wind regimes at Wels and Feldbach are

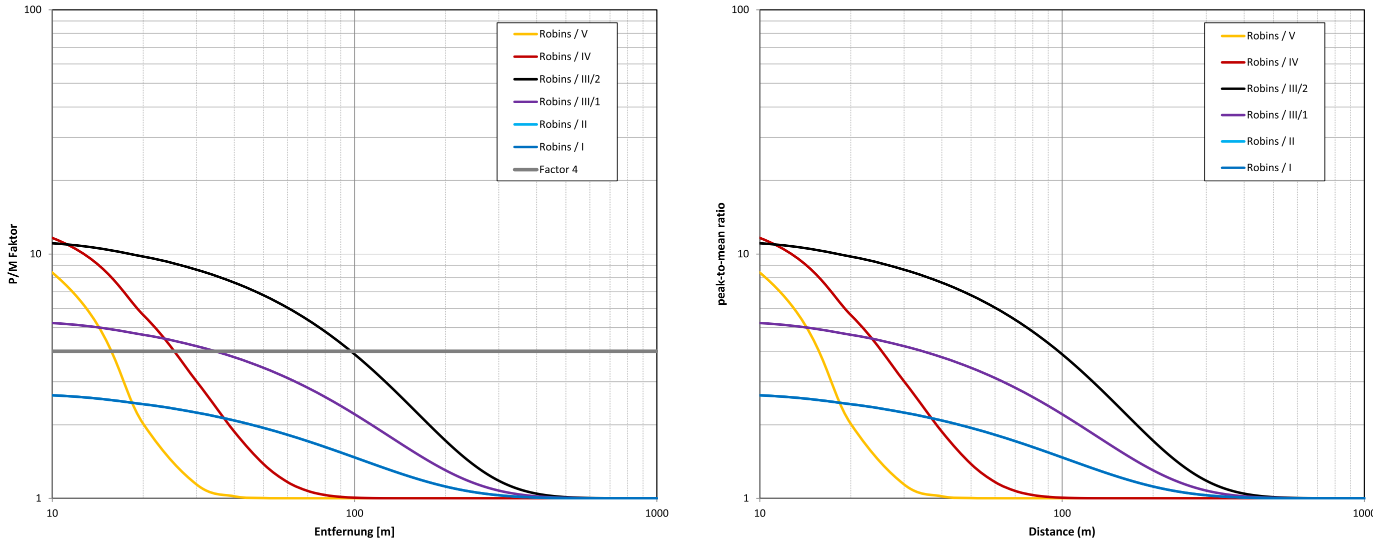

Fig. 1. Peak-to-mean ratios depending on stability classes according to the Klug/Manier scheme and the distance from the source (from Piringer et al., 2015).

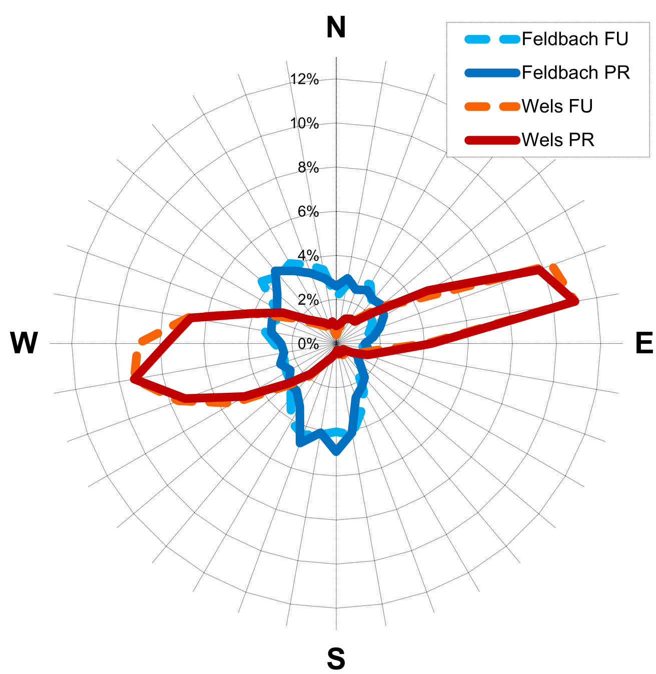

Fig. 2. Wind roses for Feldbach and Wels (PR = present (1981–2010), FU = future (2036–2065)).

very different (Figs. 2 and 3). At Wels, a site characteristic for the Austrian north-alpine foreland, we can expect a west-east orientation of the wind regime as depicted in Fig. 2. The western component is a bit deflected towards WSW, attributable to outflow from the Traun river valley. Westerly airflow in Wels is frequently connected with cyclonic activity. The easterly wind directions stand for fair-weather conditions or the flow ahead of cyclones arriving from the west. Westerly winds in Wels show on average slightly higher wind speeds than winds from east.

Feldbach is typical for valleys south of the Austrian Alps which are far less windy than the flatlands north of the Alps. The differences in the frequencies of wind directions at Feldbach are far less pronounced than at Wels, resulting in an almost circular shape of directions (Fig. 2). The mean wind speed at Feldbach is about 1 ms^{-1} lower than at Wels.

Comparing the distributions for the present and the future climate, there are almost no changes in the wind directions. As far as wind speed is concerned (Fig. 3), the future climate scenario shows an increase in daytime wind speeds at Feldbach by about 0,5 ms^{-1}; during the rest of the time and at Wels, almost no changes occur.

The average daily course of the radiation balance is as expected (Fig. 4): outgoing long-wave radiation dominates at night, whereas between sunrise and sunset, the radiation balance is on average positive, culminating at approx. 300 Wm^{-2} on average around noon at both sites. The maximum values at Feldbach are slightly higher than at Wels because the south-alpine site is positioned further south and less cloudy and windy than the area around Wels. Only at Feldbach, a slight

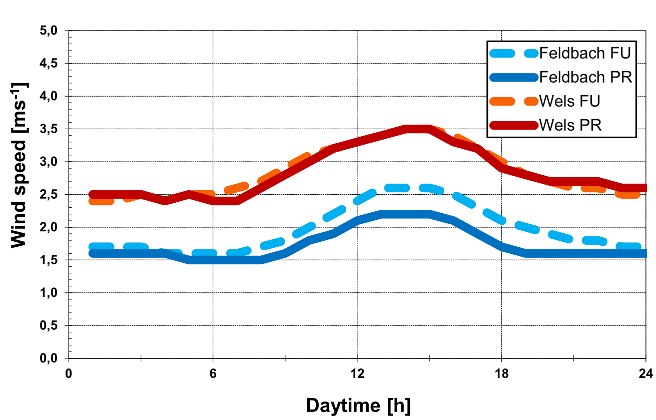

Fig. 3. Annual mean of the daily course of wind speed for Feldbach and Wels (PR = present (1981–2010), FU = future (2036–2065)).

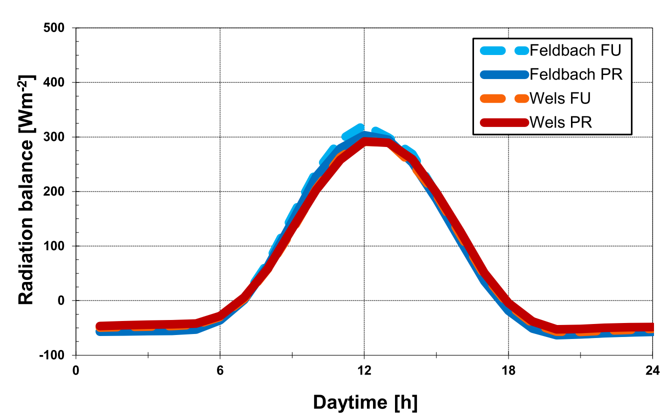

Fig. 4. Annual mean of the daily course of the radiation balance for Feldbach and Wels (PR = present (1981–2010), FU = future (2036–2065)).

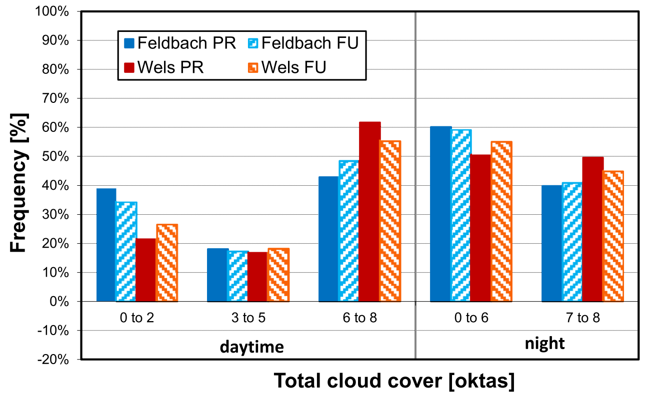

Fig. 5. Frequency distribution of total cloud cover at Feldbach and Wels (PR = present (1981–2010), FU = future (2036–2065)) at daytime and night.

increase in the radiation balance around noon is calculated for the future climate whereas no changes at Wels are shown in Fig. 4.

The frequency distribution of the total cloud cover is depicted in Fig. 5. The class separation is taken from the Klug/Manier scheme (Table 1). There are 3 classes during daytime and only 2 at night. Especially at daytime, Feldbach is less cloudy than Wels, both in the present and the future climate scenario. This agrees well with the slightly higher values of the radiation balance at Feldbach (Fig. 4). At night-time, this is also the case, but not as pronounced. The expected changes in cloudiness between the present and the future climate differ at the two locations: whereas the percentage of low cloud cover will decrease and that of high cloud cover will increase by about 5% at Feldbach, especially at daytime, it is the opposite at Wels (Fig. 5). At night, Feldbach shows almost no changes, but at Wels, the same behaviour as during daytime is predicted, i.e. lower cloud cover will increase, high cloud cover will decrease, each by about 5%.

Whereas there are only marginal changes between the present and the future climate scenarios for the parameters wind direction, wind speed, and radiation balance, a systematic increase of air temperature is observed at both sites, as expected. The annual mean of the daily course of air temperature is shown in Fig. 6. For both climate scenarios, the day-night differences are larger in Feldbach than in Wels, caused by the lower winds and clearer skies at the former. Night-time temperatures are on average higher, daytime temperatures on average lower in Wels than in Feldbach. The average temperature increase between the present and the future climate scenario is mostly around 1.0 °C at Wels and 1.5 °C at Feldbach.

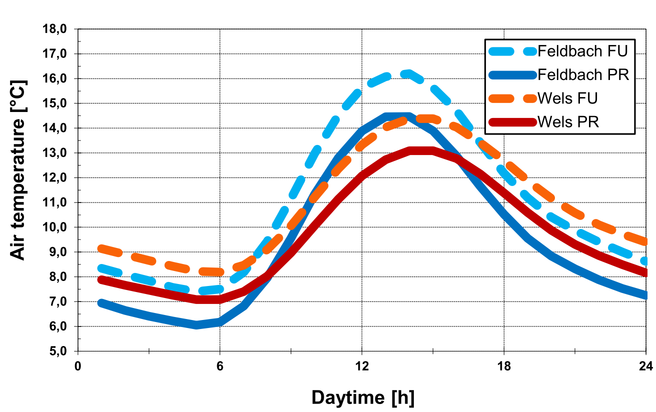

Fig. 6. Annual mean of the daily course of air temperature for Feldbach and Wels (PR = present (1981–2010), FU = future (2036–2065)).

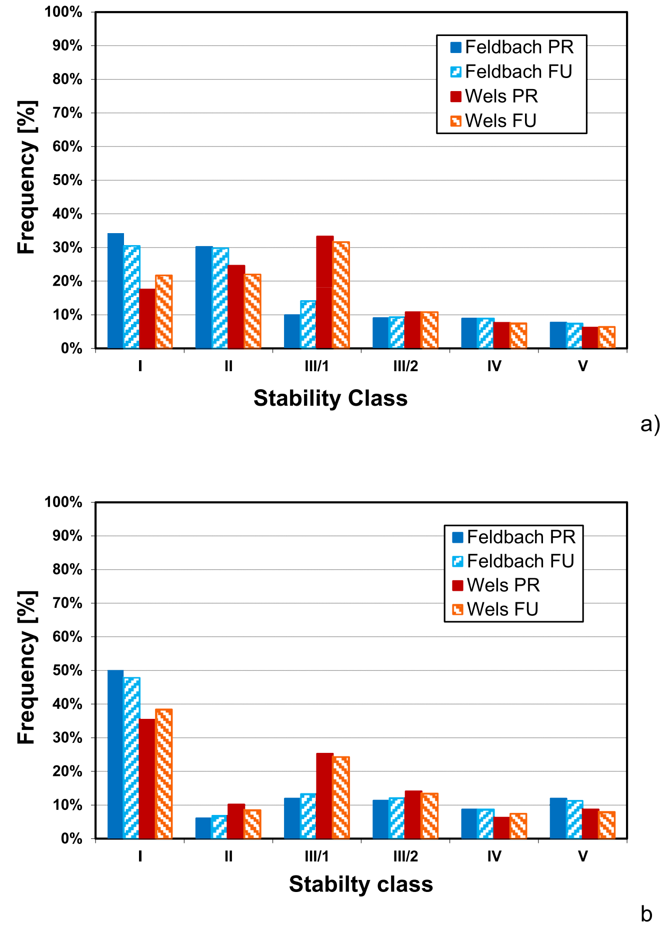

Fig. 7. Frequency distribution of stability classes determined by (a) cloudiness and wind speed (Table 1) and (b) radiation balance and wind speed (Table 2) at Feldbach and Wels (PR = present (1981–2010), FU = future (2036–2065)).

3.2. Changes in stability classes

For this investigation, two methods to determine atmospheric stability have been used (Section 2.2). The results of the different methods and the changes between the two climate scenarios are shown in Fig. 7. In the two figures, the stability classes are grouped from very stable (class I) to very unstable (class V).

There are systematic differences in the distribution of stability classes at both sites. At Wels in the present scenario, using the cloudiness method (Fig. 7a), class III/1 with more than 30% occurs most frequently (neutral at night), whereas using the radiation balance (Fig. 7b), the stable class I with 35 to almost 40% is most frequent, and this method delivers also most cases for class III/2 (neutral during daytime). In the future, both methods show that stability class I will occur slightly more frequent, with an increase of about 5%. This is the effect of the reduced cloud amount (Fig. 5). Classes II and III/1 will occur slightly less frequent, whereas no changes are seen for the other classes (towards instability).

Feldbach is generally characterized by more stable and unstable situations and less neutral conditions, compared to Wels (Fig. 7a and b). The radiation balance method dominates only in very stable conditions (class I; Fig. 7b). The cloudiness method delivers the highest frequency for classes II and I (Fig. 7a). For classes III/2, IV and V, the cloudiness and the radiation balance methods deliver very similar amounts. Comparing the present and the future scenario, the share for stability class I will be slightly reduced and that for class III/1 slightly increased at Feldbach. This is more pronounced for the cloudiness method and is a direct effect of the increase in cloud amount in Feldbach (Fig. 5).

Almost no changes occur for the other stability classes.

To summarize, the two stations show a difference in the forecasted changes. Using the cloudiness method (Fig. 7a), Feldbach shows an about 5% decrease of very stable conditions and the same increase of neutral conditions at night. At Wels, the opposite is the case: there is an increase in very stable conditions by about 5% and a slighter decrease for classes II and III/1. As in Feldbach, almost no changes in daytime stability will occur.

When using the radiation balance to determine stability classes (Fig. 7b), small changes for all classes at both sites are to be expected. Again, these changes will be different for the two stations. At Feldbach, a slight decrease in the frequency occurs for very stable (class I) and very unstable conditions (class V); for all other classes except IV, a slight increase in the frequency of stability classes is expected. At Wels, again mostly the opposite will be the case: the frequencies for classes I and IV will increase, for all others slightly decrease.

3.3. Odour impact: changes in direction-dependent separation distances

The direction depending separation distances have been calculated for the two sites, the two stability schemes and for the present and future climate. They have been calculated for two odour impact criteria according to GOAA (2008), namely for a threshold of 1 $ou_E\ m^{-3}$ and exceedance probabilities of 10% for pure residential areas and 15% for commercial/industrial and agriculturally dominated areas.

The separation distances are shown as isopleths of the OIC in Fig. 8, encompassing the area of exceedance of the given thresholds. An increase in (tolerated) exceedance probability reduces the affected area; a limit value of 15% (Fig. 8b) is thus more unfavourable for residents than a limit value of 10% (a higher level of protection, Fig. 8a). The black rectangle in the middle of both charts in Fig. 8 is the livestock building. The area displayed is 1000 × 1000 m, with the odour source in the centre. The bold lines depict separation distances obtained by the method based on the cloudiness, the thin lines those by the method based on the radiation balance (Section 2.2). Separation distances at Wels are shown in red and black/grey, those at Feldbach in blue and green. Dashed lines represent the future scenario.

In Fig. 8, the shape of the separation distances resembles to some extent the form of the wind roses (Fig. 2), especially at Wels. Generally, the separation distances at Wels are larger than at Feldbach. This can be explained mainly by the more frequent higher wind speeds at Wels which occur primarily in combination with neutral conditions (Table 1) with the highest peak-to-mean ratios (Fig. 1). At Wels, the differences in separation distances between the scenarios are generally smaller than at Feldbach, because the changes in the meteorological parameters are smaller (Section 3.1).

Starting with the separation distances obtained for an exceedance probability of 10% (Fig. 8a), an increase in separation distances between the present and the future climate is observed at Feldbach, for both methods to determine stability classes, especially towards north. This is about additional 30 m, resulting in a separation distance of 200 m north of the livestock unit. Slightly shorter distances are obtained for the radiation balance method. Towards NNE, the increases are even larger and are more than 50 m for the cloudiness method. Towards SE, the increase is smaller, especially for the cloudiness method. The maximum separation distances are then about 180 m, with again lower values for the radiation balance method.

At Wels, using the cloudiness method to determine stability, the maximum separation distances for an exceedance probability of 10% in the current climate are 320 m towards NE and 380 m towards SW. In the future, a very slight increase of maximum separation distances for 10% to 330 m towards NE and 400 m towards SW are obtained. Using the radiation balance method to determine atmospheric stability, slightly shorter maximum separation distances than when using the cloudiness method are obtained, and the changes between the scenarios are less pronounced. For the current climate and an exceedance probability of 10%, 320 m towards NE and 350 m towards SW are obtained as maximum separation distances. In the future, only little changes for maximum separation distances for an exceedance probability of 10% are observed.

As already mentioned, an increase in the tolerated exceedance probability to 15% (Fig. 8b) reduces the separation distances. The most

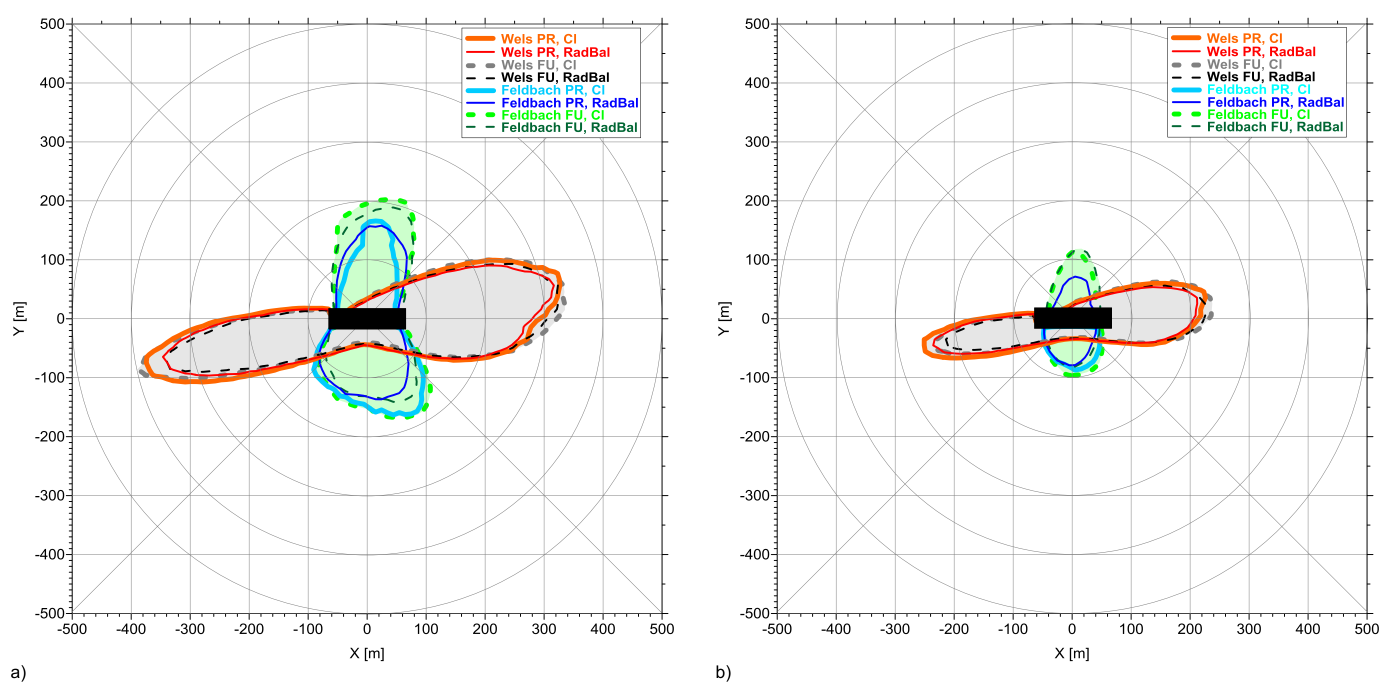

Fig. 8. Direction-dependent separation distances (m) for (a) 10% exceedance probability (pure residential areas) and for (b) 15% exceedance probability (commercial/industrial and agriculturally dominated areas) at Feldbach and Wels, both for PR (present climate (1981–2010)) and FU (future climate (2036–2065)); based on a combination of cloudiness and wind speed (Cl) and of radiation balance and wind speed (RadBal) to determine stability classes; the black rectangle is the livestock building.

significant changes occur for the cloudiness method at Feldbach. If 15% are tolerated, no separation distance north of the livestock unit can be determined in the present climate, probably caused by the combination of relatively low wind speeds (Fig. 3) and low frequencies of neutral stability (Fig. 7a) which is associated with the highest peak-to-mean factor (Fig. 1). In the future scenario, a maximum separation distance of about 100 m towards north is obtained, whereas only a small increase towards south to about 100 m occurs. Using the radiation balance to determine atmospheric stability, a separation distance of about 60 m is obtained towards the north in the current climate, increasing to about 100 m for the future climate scenario. Towards south, maximum separation distances are around 100 m, independent of the climate scenario and the method to determine atmospheric stability. Compared to an exceedance probability of 10% (Fig. 8a), a shift of the maximum separation distances from SE to S is observed.

At Wels, the separation distances are again larger, but the changes between present and future climate are smaller than at Feldbach. Compared to 10% (Fig. 8a), they are reduced to about 220 m towards NE and 250 m towards SW in the present climate (Fig. 8b); in the future, the maximum distances are about 230 m in both directions, leading to a slight increase towards easterly directions and a slight decrease towards SW.

4. Discussion

Climate change can affect air quality in a number of ways (Athanassiadou et al., 2010). For odorous substances and ammonia as a proxy for odour, the climate change signal has an impact on the emission not only by the temperature related production by bacterial activity and the release of these chemical compounds, but also by the wind velocity ((Geels et al., 2012; Schauberger et al., 2013; Schauberger et al., 2018; Skjøth and Geels, 2013; Sutton et al., 2013). In our approach we assume an annual mean value of the odour emission rate to emphasize the impact on the dispersion parameters (wind speed and direction, and the stability of the atmosphere), which are the main focus of this paper. The impact of the outdoor situation on the indoor climate of the livestock buildings (temperature, ventilation rate, animal activity) was faded out (Schauberger et al., 2014, 2018).

For some substances, the atmospheric chemistry is relevant. In general it is assumed as a working hypothesis that odorous substances behave like inert gases without chemical reactions or adsorption (Schauberger et al., 2012). There are some indications that this working hypothesis needs to be revised in some aspects (Trabue et al., 2011). For other pollutants (e.g. ammonia), the atmospheric chemistry is well elaborated. For bioaerosols, the viability of the bacteria and viruses in the atmosphere is a key factor (DasSarma and DasSarma, 2018), which is also related to climate change (van Leuken et al., 2016).

In this paper we emphasize on the climatic changes of the meteorological parameters necessary to run the Lagrangian particle diffusion model LASAT to calculate direction-dependent separation distances around a fictitious, but typical livestock unit at two Austrian sites situated in areas with considerable livestock activity, however characterized by different climatological conditions. Wels in the North-alpine foreland shows main wind directions from south-west and north-east, a good ventilation and a tendency to neutral stability. Feldbach in the Raab valley south of the main Alpine chain experiences frequent calm wind conditions and a large abundance of stable conditions. At both sites, the same livestock building was assumed, and the same climate periods, 1981–2010 for the present climate and 2036–2065 for the future climate, were applied. For both sites and for both climate scenarios, the two odour impact criteria OIC were used to determine the separation distances, namely those of the German GOAA (2008). Both use a threshold of 1 ou_E m^{-3} but different exceedance probabilities: 10% for residential and 15% for commercial/industrial areas. This guideline is widely applied in Germany and Austria. Beside this German odour impact criterion, many other national OIC are in use (Brancher et al., 2017) but they result in similar protection distances (Sommer-Quabach et al., 2014).

The current investigation shows, apart from the expected increase in air temperature (Fig. 6), a varying degree of changes of the input parameters to the dispersion model between the present and the future climate scenarios. The smallest changes occur for wind direction (Fig. 2) and radiation balance (Fig. 4). More relevant – and also different between the sites – are the changes in wind speed (Fig. 3) and cloud cover (Fig. 5) which affect directly the atmospheric stability (Fig. 7). The south-alpine site Feldbach shows an increase in daytime wind speed and an increase in cloud cover. These changes result in a reduction of very stable and an increase in neutral conditions, especially for the cloud amount method (Fig. 7a). Very stable conditions are defined for clear skies and low wind speeds at night, neutral conditions for higher wind speeds and larger cloud amounts, both day and night. Hence, an increase in wind speed and an increase in cloudiness can explain the observed changes in atmospheric stability. Especially for the cloud amount method to determine stability, changes up to 5% in very stable and neutral dispersion categories have to be expected.

At Wels in the north-alpine foreland of Austria, no changes in wind speed are observed (Fig. 3), but the cloud amount will change at a similar rate of around 5% as in Feldbach, but it will decrease, both day and night (Fig. 5). The resulting stability classes will change accordingly: very stable conditions will increase, slightly stable and neutral conditions will decrease, for both methods to determine atmospheric stability. Only small changes are observed for the other stability classes. These contrasting results for the two investigated sites show that the selected climate scenarios are sensitive to the very different local meteorological conditions at Feldbach and Wels. Feldbach experiences much lower wind speeds than Wels, although when they will increase in the future; the site is also characterized by more stable conditions than Wels, even when they will decrease in the future.

The resulting lengths and the changes in separation distances at the two sites can also be explained quite straightforward, based on the analysis of the meteorological data. The separation distances have been determined for the two odour impact criteria according to GOAA (2008), namely a threshold of 1 ou_E m^{-3} and tolerated exceedance probabilities of 10% for residential or 15% for commercial/industrial and agricultural areas, with the latter being more unfavourable for the neighbourhood. An increase in separation distances means that the area of neighbourhood protection also increases. Because of the higher wind speeds and the predominantly neutral stability conditions with the highest peak-to-mean ratios (Fig. 1), the maximum separation distances at Wels are more than two times larger than at Feldbach (Fig. 8a and b). Separation distances have been calculated for the same odour emission rate, at both sites. The changes in separation distances between the present and the future climate can be explained directly by the changes in the meteorological conditions: there is a tendency that maximum separation distances will slightly increase in Feldbach, due to the increase in wind speed and neutral stability classes; there is a mixed result for Wels and only small changes, but the maximum separation distances towards south-west will be reduced in the future, especially for an exceedance probability of 10% for residential areas (Fig. 8a). The most significant changes occur for the cloudiness method at Feldbach. If 15% are tolerated, no separation distance north of the livestock unit can be determined in the present climate, probably caused by the combination of relatively low wind speeds (Fig. 3) and low frequencies of neutral stability (Fig. 7a) which is associated with the highest peak-to-mean factor (Fig. 1). In the future scenario, a maximum separation distance of about 100 m towards north is obtained.

These small modifications of the dispersion parameters are in good agreement to the air stagnation index for Central Europe which is predicted for the near future by Horton et al. (2014). Athanassiadou et al. (2010) investigated the climate change impact on the dispersion parameters and the related ambient concentration for the UK with similar results for small sources. The ambient concentration is changing less than 10% not only for the mean value but also for several percentiles. Doherty et al. (2017) concluded that the impact of climate change on air quality and the related human health will be dominated by emission changes rather than changes of the dispersion of pollutants in the atmosphere. This can also be expected for odour and the related separation distances.

5. Conclusions

The main focus of this investigation is the impact of the changing climate on the dilution of airborne emissions. This was demonstrated by the emission of odorous substances from a livestock building. Based on the ambient odour concentration, calculated by a dispersion model, the separation distances around this livestock farm were determined to protect the neighbourhood from excessive annoyance. A considerable increase in these distances would exert additional pressure on livestock farming, apart from possible losses from increased indoor heat stress due to rising air temperatures (Mikovits et al., 2018).

Separation distances have been calculated with the Lagrangian particle dispersion model LASAT, applying the peak-to-mean approach by Piringer et al. (2015) to account for the ability to detect odour in the time scale of a single human breath (approx. 5 s). We compiled two climate data sets, one for the current climate (1981–2010) and one for the period 2036–2065 (future climate) at two sites representative for livestock husbandry in Austria but discernible in average climate conditions, the area of Wels in the north-alpine foreland and the Raab valley at Feldbach south of the main Alpine chain. For all scenarios, the same source data have been used (Table 3). Separation distances have been calculated for two odour impact criteria OIC according to GOAA (2008), namely 10 and 15% exceedance probability, each in combination with an odour threshold of 1 ou_E m^{-3}.

In general, only small changes in the input parameters to the dispersion model have been calculated for the two climate scenarios (Sections 3.1 and 3.2). This leads nevertheless to differences of the changes of separation distances at the two sites investigated (Fig. 8a and b):

The changes in separation distances between the current and the future climate scenarios are larger at Feldbach than at Wels. The most significant change is seen at Feldbach when using the cloudiness method to determine stability for an exceedance probability of 15% north of the livestock unit: in the present climate, no separation distance north of the livestock unit can be determined; in the future scenario, a maximum separation distance of about 100 m towards north is obtained (Fig. 8b), probably caused by the combination of relatively low wind speeds (Fig. 3) and low frequencies of neutral stability (Fig. 7a) which is associated with the highest peak-to-mean factor (Fig. 1). Generally at Feldbach, a slight increase in separation distances from the present to the future climate has to be expected, especially when using the cloudiness method. At Wels, only small changes in separation distances are predicted, with a tendency of a slight reduction of the largest separation distance when using the cloudiness method (for an exceedance probability of 10%) and varying changes when using the radiation balance method.

These results demonstrate the relevance of the investigation. Local meteorology exerts a profound impact on a site-specific quantity like the separation distance to protect the neighbourhood from odour annoyance. Because of the relatively complex interaction of the meteorological parameters and the peak-to-mean factors when determining separation distances for given exceedance probabilities, no a priori forecast is possible. In a region of interest, separation distances will increase or decrease in a future climate scenario. A decrease will certainly lead to a relief for planning and zoning purposes. If the outcome of such an investigation is an increase like in Feldbach, practice will show in the future which increases will be tolerated and which will cause technical or legal efforts to overcome these unwanted changes. Due to the durability of today's investment decisions for new livestock facilities, location-sensitive results as presented in this study can be valuable tools for land use planning and zoning.

Declaration of interests

I declare that I want to inform the scientific community about my latest research.

Acknowledgements

The project *PiPoCooL Climate change and future pig and poultry production: implications for animal health, welfare, performance, environment and economic consequences* is funded by the Austrian Climate and Energy Fund in the framework of the Austrian Climate Research Program (ACRP8 – PiPoCooL – KR15AC8K12646).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2019.100021.

References

Athanassiadou, M., Baker, J., Carruthers, D., Collins, W., Girnary, S., Hassell, D., Hort, M., Johnson, C., Johnson, K., Jones, R., Thomson, D., Trought, N., Witham, C., 2010. An assessment of the impact of climate change on air quality at two UK sites. Atmos. Environ. 44, 1877–1886.

Baumann-Stanzer, K., Piringer, M., Polreich, E., Hirtl, M., Petz, E., Bügelmayer, M., 2008. User experience with model validation exercises. Ext. abstract of the 12th Int. Conf. on harmonization within atmospheric dispersion modeling for regulatory purposes, Cavtat, Croatia, 6 – 10 Oct. 2008. Croat. Meteorol. J. 43 (1), 52–56 ISSN 1330-0083.

Baumann-Stanzer, K., Leitl, B., Trini-Castelli, S., Millies, M., Rau, G., all COST ES1006 members, 2014. Local-scale modelling of accidental releases in built environments - selected results of the Michelstadt" model evaluation exercise in COST Action ES1006. In: Proc. 9th Int. Conf. On Air Quality - Science and Application, Garmisch-Partenkirchen, 24 - 28 March 2014, pp. 106.

Brancher, M., Griffiths, K.D., Franco, D., De Melo Lisboa, H., 2017. A review of odour impact criteria in selected countries around the world. Chemosphere 168, 1531–1570.

Chimani, B., Heinrich, G., Hofstätter, M., Kerschbaumer, M., Kienberger, S., Leuprecht, A., Lexer, A., Peßenteiner, S., Poetsch, M.S., Salzmann, M., Spiekermann, R., Switanek, M., Truhetz, H., 2016. ÖKS15 - Klimaszenarien für Österreich. Daten, Methoden und Klimaanalyse. Projektendbericht, Wien.

DasSarma, P., DasSarma, S., 2018. Survival of microbes in Earth's stratosphere. Curr. Opin. Microbiol. 43, 24–30.

Dee, D.P., Uppala, S.M., Simmons, A.J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M.A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A.C.M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A.J., Haimberger, L., Healy, S.B., Hersbach, H., Hólm, E.V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A.P., Monge-Sanz, B.M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., Vitart, F., 2011. The ERA-Interim reanalysis: configuration and performance of the data assimilation system. Q. J. R. Meteorol. Soc. 137, 553–597. https://doi.org/10.1002/qj.828.

Doherty, R.M., Heal, M.R., O'Connor, F.M., 2017. Climate change impacts on human health over Europe through its effect on air quality. Environ. Health Global Access Sci. Sour. 16.

Geels, C., Skjøth, C.A., Christensen, J.H., Ellermann, T., Løfstrøm, P., Gyldenkærne, S., Brandt, J., Hansen, K., Frohn, L., Hertel, O., 2012. Improved modelling of atmospheric ammonia over Denmark using the coupled modelling system DAMOS. Biogeosciences 9, 2625.

GOAA, 2008. Guideline on Odour in Ambient Air. Detection and Assessment of Odour in Ambient Air. Second Version, Berlin, Germany.

Hirtl, M., Baumann-Stanzer, K., 2007. Evaluation of two dispersion models (ADMS-Roads and LASAT) applied to street canyons in Stockholm, London and Berlin. Atmos. Environ. 41, 5959–5971.

Hirtl, M., Baumann-Stanzer, K., Kaiser, A., Petz, E., Rau, G., 2007. Evaluation of three dispersion models for the Trbovlje power plant, Slovenia. In: Carruthers, D.J., McHugh, C.A. (Eds.), Proceedings of the 11th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes; Held in Cambridge, UK, July 2e5, 2007, 21e25.

Horton, D.E., Skinner, C.B., Singh, D., Diffenbaugh, N.S., 2014. Occurrence and persistence of future atmospheric stagnation events. Nat. Clim. Change 4, 698.

Hou, P., Wu, S., 2016. Long-term changes in extreme air pollution meteorology and the implications for air quality. Sci. Rep. 6.

Huang, Q., Cai, X., Song, Y., Zhu, T., 2017. Air stagnation in China (1985-2014): climatological mean features and trends. Atmos. Chem. Phys. 17, 7793–7805.

Jacob, D., Petersen, J., Eggert, B., Alias, A., Bøssing Christensen, O., Bouwer, L.M., Braun, A., Colette, A., Déqué, M., Georgievski, G., Georgopoulou, E., Gobiet, A., Menut, L., Nikulin, G., Haensler, A., Hempelmann, N., Jones, C., Keuler, K., Kovats, S., Kröner, N., Kotlarski, S., Kriegsmann, A., Martin, E., Meijgaard, E., Moseley, C., Pfeifer, S., Preuschmann, S., Radermacher, C., Radtke, K., Rechid, D., Rounsevell, M., Samuelsson, P., Somot, S., Soussana, J.-F., Teichmann, C., Valentini, R., Vautard, R., Weber, B., Yiou, P., 2014. EURO-CORDEX: new high-resolution climate change projections for European impact research. Reg. Environ. Change 14 (2), 563.

Janicke Consulting, 2013. Dispersion Model LASAT. Reference/Working Book for Version 3.3.

Kottek, M., Grieser, J., Beck, C., Rudolf, B., Rubel, F., 2006. World map of the Köppen-Geiger climate classification updated. Meteorol. Z. 15 (3), 259–263.

KTA 1508, 2006. Instrumentierung zur Ermittlung der Ausbreitung radioaktiver Stoffe in der Atmosphäre. Sicherheitstechnische Regel des KTA Fassung 11/06.

Leelőssy, Á., Molnár Jr., F., Izsák, F., Havasi, Á., Lagzi, I., Mészáros, R., 2014. Dispersion modeling of air pollutants in the atmosphere: a review. Cent. Eur. J. Geosci. 6, 257–278.

Mikovits, C., Zollitsch, W., Hörtenhuber, S.J., Baumgartner, J., Niebuhr, K., Piringer, M., Anders, I., Andre, K., Hennig-Pauka, I., Schönhart, M., Schauberger, G., 2018. Impacts of global warming on confined livestock systems for growing-fattening pigs: simulation of heat stress for 1981 to 2017 in Central Europe. Int. J. Biometeorol (in press).

Piringer, M., Baumann-Stanzer, K., 2009. Selected results of a model validation exercise. Adv. Sci. Res. 3, 13–16.

Piringer, M., Joffre, S. (Eds.), 2005. The Urban Surface Energy Budget and Mixing Height in European Cities: Data, Models and Challenges for Urban Meteorology and Air quality.Final Report of Working Group 2 of COST Action 715. Demetra Ltd. Publishers, Printed in Bulgaria ISBN 954-9526-29-1, 239 pp.

Piringer, M., Schauberger, G., 2013. Dispersion modeling for odour exposure assessment. In: Belgiorno, V., Naddeo, V., Zarra, T. (Eds.), Odour Impact Assessment Handbook. Wiley, pp. 125–174 ISBN: 978-1-119-96928-0.

Piringer, M., Petz, E., Groehn, I., Schauberger, G., 2007. A sensitivity study of separation distances calculated with the Austrian Odour Dispersion Model (AODM). Atmos. Environ. 41, 1725–1735.

Piringer, M., Knauder, W., Petz, E., Schauberger, G., 2015. A comparison of separation distances against odour annoyance calculated with two models. Atmos. Environ. 116, 22–35.

Rockel, B., Will, A., Hense, A., 2008. The regional climate model COSMO-CLM (CCLM). Meteorol. Z. 17 (4), 347–348 2008.

Schatzmann, M., Olesen, H., Franke, J. (Eds.), 2010. COST 732 Model Evaluation Case Studies: Approach and Results. COST Action 732 - Quality Assurance and Improvement of Microscale Meteorological Models, 121 pp.

Schauberger, G., Piringer, M., Petz, E., 2000. Diurnal and annual variation of the sensation distance of odour emitted by livestock buildings calculated by the Austrian odour dispersion model (AODM). Atmos. Environ. 34, 4839–4851.

Schauberger, G., Piringer, M., Petz, E., 2002. Calculating direction-dependent separation distance by a dispersion model to avoid livestock odour annoyance. Biosyst. Eng. 82, 25–37.

Schauberger, G., Piringer, M., Knauder, W., Petz, E., 2011. Odour emissions from a waste treatment plant using an inverse dispersion technique. Atmos. Environ. 45, 1639–1647.

Schauberger, G., Piringer, M., Schmitzer, R., Kamp, M., Sowa, A., Koch, R., Eckhof, W., Grimm, E., Kypke, J., Hartung, E., 2012. Concept to assess the human perception of odour by estimating short-time peak concentrations from one-hour mean values. Atmos. Environ. 54, 624–628.

Schauberger, G., Lim, T.T., Ni, J.Q., Bundy, D.S., Haymore, B.L., Diehl, C.A., Duggirala, R.K., Heber, A.J., 2013. Empirical model of odor emission from deep-pit swine finishing barns to derive a standardized odor emission factor. Atmos. Environ. 66, 84–90.

Schauberger, G., Piringer, M., Heber, A.J., 2014. Odour emission scenarios for fattening pigs as input for dispersion models: a step from an annual mean value to time series. Agric. Ecosyst. Environ. 193, 108–116.

Schauberger, G., Piringer, M., Mikovits, C., Zollitsch, W., Hörtenhuber, S.J., Baumgartner, J., Niebuhr, K., Anders, I., Andre, K., Hennig-Pauka, I., Schönhart, M., 2018. Impact of global warming on the odour and ammonia emissions of livestock buildings used for fattening pigs. Biosyst. Eng. 175, 106–114.

Skjøth, C.A., Geels, C., 2013. The effect of climate and climate change on ammonia emissions in Europe. Atmos. Chem. Phys. 13, 117–128.

Smiatek, G., Kunstmann, H., Senatore, A., 2016. EURO-CORDEX regional climate model analysis for the Greater Alpine Region: performance and expected future change. Journal of Geophysical Research. Atmosphere 121, 7710–7728. https://doi.org/10.1002/2015JD024727.

Sommer-Quabach, E., Piringer, M., Petz, E., Schauberger, G., 2014. Comparability of separation distances between odour sources and residential areas determined by various national odour impact criteria. Atmos. Environ. 95, 20–28.

Sutton, M.A., Reis, S., Riddick, S.N., Dragosits, U., Nemitz, E., Theobald, M.R., Tang, Y.S., Braban, C.F., Vieno, M., Dore, A.J., Mitchell, R.F., Wanless, S., Daunt, F., Fowler, D., Blackall, T.D., Milford, C., Flechard, C.R., Loubet, B., Massad, R., Cellier, P., Personne, E., Coheur, P.F., Clarisse, L., Van Damme, M., Ngadi, Y., Clerbaux, C., Skjøth, C.A., Geels, C., Hertel, O., Kruit, R.J.W., Pinder, R.W., Bash, J.O., Walker, J.T., Simpson, D., Horváth, L., Misselbrook, T.H., Bleeker, A., Dentener, F., de Vries, W., 2013. Towards a climate-dependent paradigm of ammonia emission and deposition. Phil. Trans. Biol. Sci. 368.

TA Luft, 2002. Erste Allgemeine Verwaltungsvorschrift zum Bundes–Immissionsschutzgesetz. (Technische Anleitung zur Reinhaltung der Luft – TA Luft). Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit.

Trabue, S., Scoggin, K., McConnell, L., Maghirang, R., Razote, E., Hatfield, J., 2011. Identifying and tracking key odorants from cattle feedlots. Atmos. Environ. 45, 4243–4251.

van Leuken, J.P.G., Swart, A.N., Droogers, P., van Pul, A., Heederik, D., Havelaar, A.H., 2016. Climate change effects on airborne pathogenic bioaerosol concentrations: a scenario analysis. Aerobiologia 32, 607–617.

VDI 3782 Part 1, 2009. Environmental Meteorology- Atmospheric Dispersion Models - Gaussian Plume Model for the Determination of Ambient Air Characteristics. Verein Deutscher Ingenieure, VDI Verlag, Düsseldorf.

VDI 3894 Part 1, 2011. Emissions and Immissions from Animal Husbandry - Housing Systems and Emissions - Pigs, Cattle, Poultry, Horses. Verein Deutscher Ingenieure, VDI Verlag, Düsseldorf.

Zlatev, Z., Havasi, Á., Faragó, I., 2011. Influence of climatic changes on pollution levels in Hungary and surrounding countries. Atmosphere 2, 201–221.