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# Dry hopping potential of Eureka!, a new hop variety

To monitor the dry hopping potential of the new hop variety Eureka!, brewing trials were performed and observed in detail. Static dry hopping was done for 1, 2, 4, and 8 days. Beers were analyzed using published methods to have an insight into the transfer of 4-MMP (GC×GC-TOFMS, published by *Reglitz et al.* in *BrewingScience* 71, 2018), selected hop aroma compounds (EBC 9.49) and hop derived bitter substances (EBC 9.47). Major transfer of 4-MMP and the aroma compounds myrcene, linalool, geraniol, and 2-methylbutyl isobutanoate happened during the first two days of dry hopping with only slight further increase between days 2 and 8. The same behavior could be found for the bitter components alpha-acids and humulinones, whereas xanthohumol increased until day 4. All results of dry-hopped beers allowed a comparison with a lab scale cold-water extraction which was tested as a possible prediction tool for dry hopping of a selected hop variety. In addition to analytics, sensory evaluations were carried out for the beers with help of descriptive analysis. With focus on fruity, especially black currant-like, aroma in beer, the received findings suggest a hop contact time of 2 days of dry hopping with the hop variety Eureka!.

Descriptors: dry hopping, 4-mercapto-4-methylpentan-2-one (4-MMP), hop aroma components, hop bitter compounds, brewing trials, cold-water extraction

## 1 Introduction

A multitude of hop varieties are present on the market which are traditionally classified as aroma or bitter hops. However, all hop varieties contain compounds that are contributing to aroma and bitterness of beer (hop essential oils and hop bitter acids).

The hop variety Eureka! originated from a cross between Apollo and a male developed in the Hopsteiner breeding program. Maturing in late-season, its average yield ranges from 2,800 to more than 3,000 kg/ha. The Eureka! hops offer both, a high total hop oil content (up to 4 mL/100 g) as well as a high content of bitter components (alpha-acids between 17 and 20 % and beta-acids up to 6 %) (Table 1).

A detailed aroma evaluation of the smell of raw hops of Eureka! gave resinous, strong herbal, spicy, and fruity notes. Even though the fruity notes are less distinct, they could be specified from tropical-citrus to dark stone fruit with a special attention on black currant aroma impression (Fig. 1, [1]).

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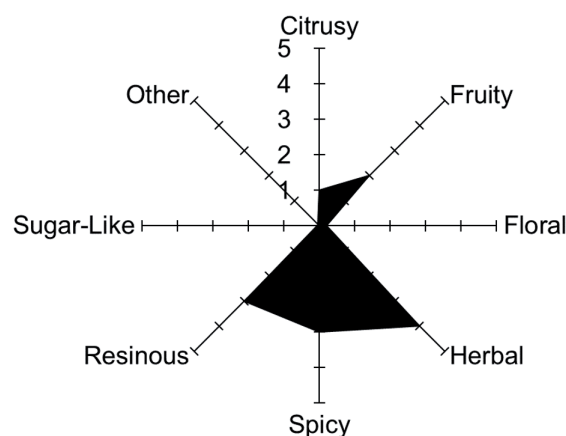
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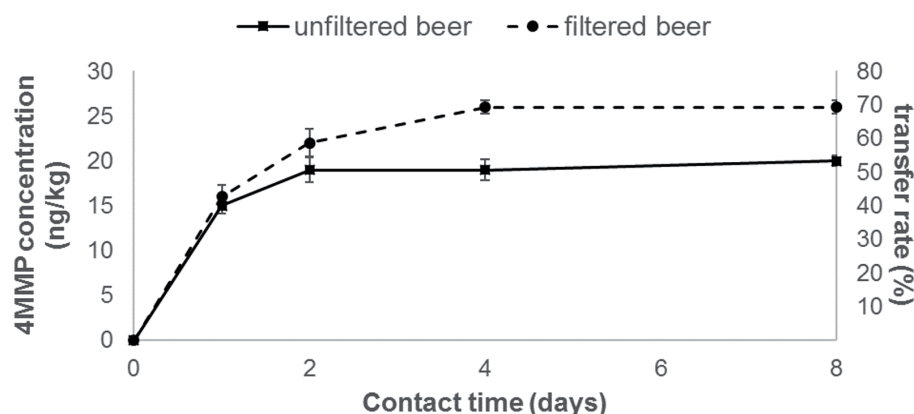
The black currant-like smelling hop odorant 4-mercapto-4-methylpentan-2-one (4-MMP) also known as 4-methyl-4-sulfanylpentan-2-one (4-MSP) was described for the first time in Cascade hops [2, 3]. In 2017, *Steinhaus* and *Reglitz* studied the influence of variety, provenance, harvest year, and processing on 4-MMP concentra-

**Table 1** Chemical ingredients of hop variety Eureka! [1]

| Components            |           |
|-----------------------|-----------|
| Alpha-acids in %      | 17.0–20.0 |
| Beta-acids in %       | 4.5–6.0   |
| Co-Humulone % rel.    | 28–30     |
| Xanthohumol           | 0.5–0.6   |
| Total Oil in mL/100 g | 2.5–4.5   |



**Fig. 1** Aroma evaluation of Eureka! (rated by smell of raw hops on a 0-5 scale)



**Fig. 2** Transfer of 4-MMP from hops into beer during dry hopping with Eureka!, published in [5] (Error bars represent  $\pm$  standard deviation of triplicate analyses)

tions [4]. To get the exact amount of this thiol, the authors developed a stable isotope dilution assay, with selective isolation of thiols by mercurated agarose and in combination with GC $\times$ GC-TOFMS and applied the method to 53 different hop samples. The value of 4-MMP in this study ranged from <1 to 114  $\mu$ g/kg. High concentrations could be found in hop varieties from the United States, with the highest level in Citra (114  $\mu$ g/kg) followed by Eureka! (59.1  $\mu$ g/kg), Simcoe (51.2  $\mu$ g/kg) and Apollo (28.6  $\mu$ g/kg). 4-MMP was absent from traditional German and English varieties. The study also showed that hop processing such as drying and pelletizing had only a minor impact on this compound but the harvest year as well as the storage influenced the amount. In the next step, the developed analytical method [4] was also used by Reglitz et al. to study the behavior of this hop odorant during dry hopping with the hop variety Eureka! [5]. Therefore, the transfer of 4-MMP was observed during static dry hopping. The results (Fig. 2) showed that the major transfer happened during the first two days of dry hopping, whereas only slight further increase between days 2 and 8 was detected [5].

Even if 4-MMP is an important hop odorant with a very low odor threshold in beer (0.5–1.5 ng/L [6]), there are other relevant key hop derived aroma compounds in beer. It is known from literature that linalool is responsible for the citrusy and floral aroma impressions in late hopped beers [7, 8]. It is also present in high concentrations in dry-hopped beers. For the monoterpene myrcene an aroma contribution to dry-hopped beer could also be observed [7]. Like 4-MMP, the terpene alcohol geraniol with an odor threshold of 4  $\mu$ g/L [2] is described to be more varietal specific than linalool [3]. The ester 2-methylbutyl 2-methylpropanoate, also known as 2-methylbutyl isobutanoate, was identified by *Takoi* et al. in 2009 as a specific flavor compound derived from Nelson Sauvin hop with an odor threshold of 78  $\mu$ g/L in beer imparting a green apple-like and/or apricot-like flavor [9].

Besides hop derived aroma compounds, many other hop ingredients dissolve into beer when the dry hopping technique is used. Isohumulones are known to be the main contributors to beer bitterness. But in the last years research was focused on the identification, quantitation, and the evaluation of the bitterness intensity of oxidized hop acids like humulinones [10–12]. *Algazzali* and *Shellhammer* reported in 2016 that humulinones have a relative bitterness intensity of 66 ( $\pm$  13) % in comparison to isohumulones [12]. Depending

on the hop variety, hops may contain up to a maximum of 1 % of the prenylflavonoid xanthohumol. The natural yellow substance was discovered more than 100 years ago. The ratio of xanthohumol to alpha-acids serves as a characteristic of each variety. Xanthohumol shows a very mild bitterness [13]. Prenylflavonoids continue to draw attention of more and more research groups. Several positive physiological and pharmacological properties have been reported for xanthohumol and other prenylflavonoids from hops [14–17].

The aim of the present work was to study the dry hopping potential of the new hop variety Eureka!. Therefore, brewing trials were carried

out and several hop derived aroma and bitter compounds were evaluated in the final beers with help of analytical methods together with a sensory approach. In addition, a lab scale cold-water extraction method was performed, and the corresponding results were compared to the findings of the real brewing trials. In this work, the same beers were analyzed as published recently by Reglitz et al. (2018) where the authors focused on 4-MMP only [5].

## 2 Materials and methods

### 2.1 Beer samples

The base beer for the dry hopping experiments was a German Pilsner type beer. The hopping regime was a single hop addition at the beginning of wort boiling with pellets of the varieties Perle and Hallertauer Tradition as well as ethanol extract made of Herkules, Hallertauer Magnum and Hallertauer Taurus. The beer was centrifuged but not filtered. The analytical parameters of the beer were as followed: 11.6 % original gravity, 5.0 vol% Ethanol, pH 4.5, and 35 International Bitter Units (IBU). The dry hopping procedure was done side by side in four cylindroconical vessels (10 hL) at  $-1$  °C. Each vessel contained the base beer from one batch. The pellets type 90 of US Eureka! hops (harvest 2016) were added from the top of the vessels and the total amount was 250 g hops/hL beer. The dry hopping was done in a static way. After the contact time of 24 h, the hop sediment was removed from the bottom of the first vessel. In the same way, vessel 2 (after 2 days), vessel 3 (after 4 days), and vessel 4 (after 8 days) were treated. Each vessel was separated into 2 parts and one part was filtered using diatomaceous earth (100 g/hL) as filter aid. Filtered and unfiltered beers were bottled after air removal and closed with crown corks with oxygen scavenging properties. The bottles were stored at 4 °C. The finished beers were analyzed direct after the brewing trials.

### 2.2 Lab scale cold-water extraction

The hop amount used in the brewing trial was 250 g/hL. Therefore, the same hop amount as well as the same batch of hop pellets type 90 of US Eureka! (harvest 2016) were used for the lab scale method. The extraction solvent was a 5 % ethanol (v/v) buffered water with a pH 4.5. Citrate buffer (0.05 M) was used and the pH was adjusted with 45 % (w/w) potassium hydroxide solution.

1 L scale was carried out. The temperature of the ethanolic buffered water was 20 °C. The extraction was done for 24 h with only a moderate agitation using a lab shaker. After 24 h, the whole hop solution was filtered using a folded filter (typical retention > 20 µm). The extraction procedure was done in triplicate.

### 2.3 Determination of 4-mercapto-4-methylpentan-2-one (4-MMP) by GC×GC-TOFMS

For the 4-MMP quantitation in cold-water extracts, a GC×GC-TOFMS method was applied [5]. The method includes the use of a stable isotope labeled standard, selective isolation of thiols by mercurated agarose and finally the GC×GC-TOFMS analysis.

### 2.4 Analysis of hop derived bitter compounds in beer by HPLC-UV

For the analysis of alpha-acids, humulinones as well as of xanthohumol in dry-hopped beers the published Analytica EBC 9.47 method was used. Method description gives [18]. Calibration standards used (Labor Veritas AG, Switzerland):

- ICE-4 for alpha-acids (Detection wavelength: 270 nm)
- ICS-Hum1 for humulinones (Detection wavelength: 270 nm)
- ICS-X1 for xanthohumol (Detection wavelength: 370 nm)

### 2.5 Determination of myrcene, linalool, geraniol, and 2-methylbutyl isobutanoate in beer by HS-Trap GC-MS

The 4 hop derived aroma components were analyzed with help of the recently published Analytica EBC 9.49 method (Hop aroma components in beer by Headspace-Trap Gas Chromatography (HT-GC)) [19].

### 2.6 Descriptive analyses of beer samples

Sensory evaluation of filtered beers was carried out using a descriptive analysis of the attributes citrusy, fruity, floral, herbal, spicy, resinous, sugar-like, and other on a 0-5 scale. Each individual category includes several sub-divisions for a more detailed specification [20]. In addition to sub-divisions given in [20], black currant-like impression was added to the fruity attribute. Further sub-divisions can be added by the panelists if it is necessary to

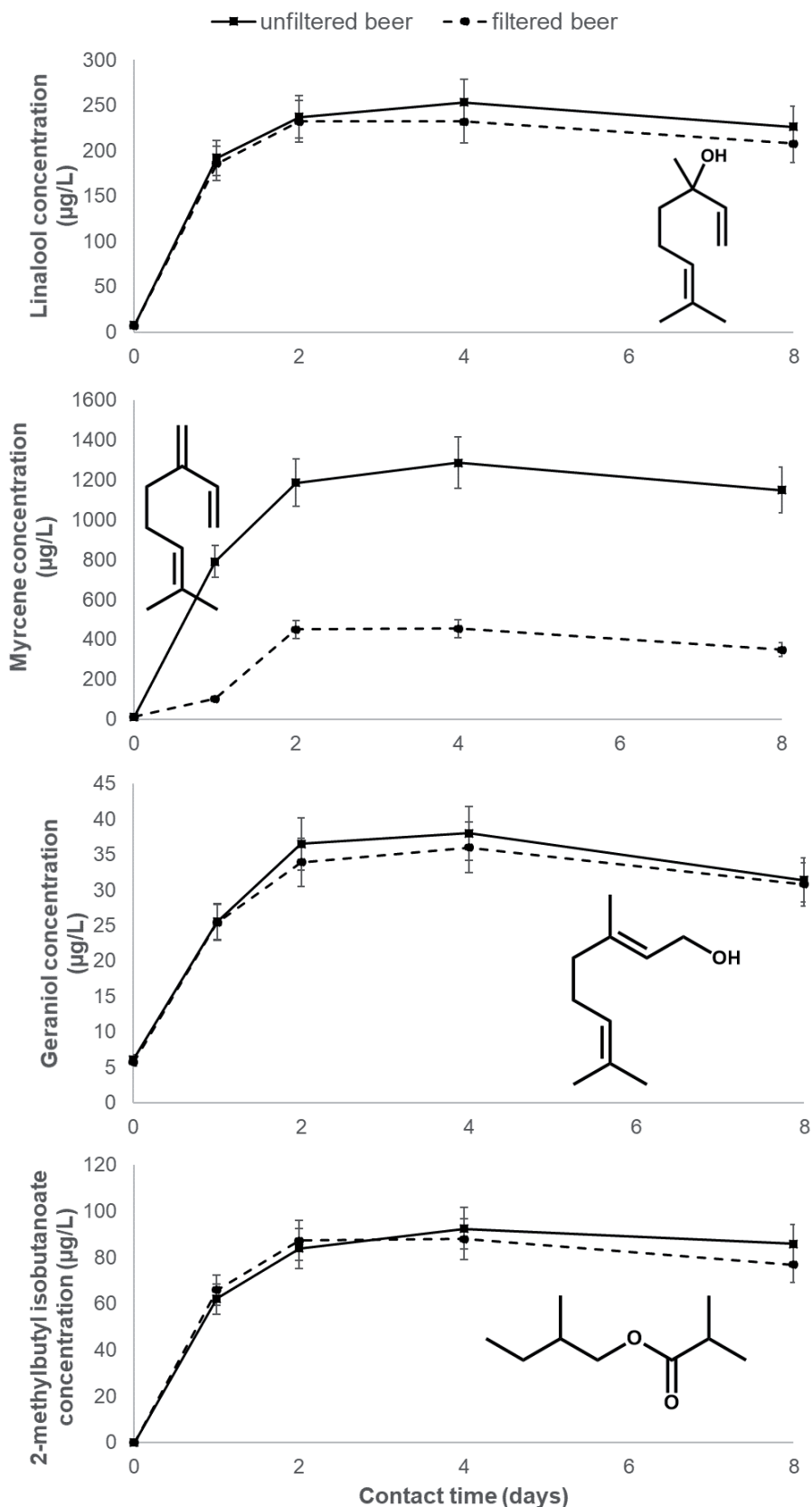


Fig. 3 Hop derived aroma components (in µg/L ± standard deviation of duplicate analyses) in unfiltered and filtered beers (base beers and dry-hopped beers with different contact time)

describe the hop aroma of a dry-hopped beer in detail. The panel for evaluation of the base beer, the beer dry-hopped for 1 day and the beer dry-hopped for 8 days consisted of 35 individuals. The

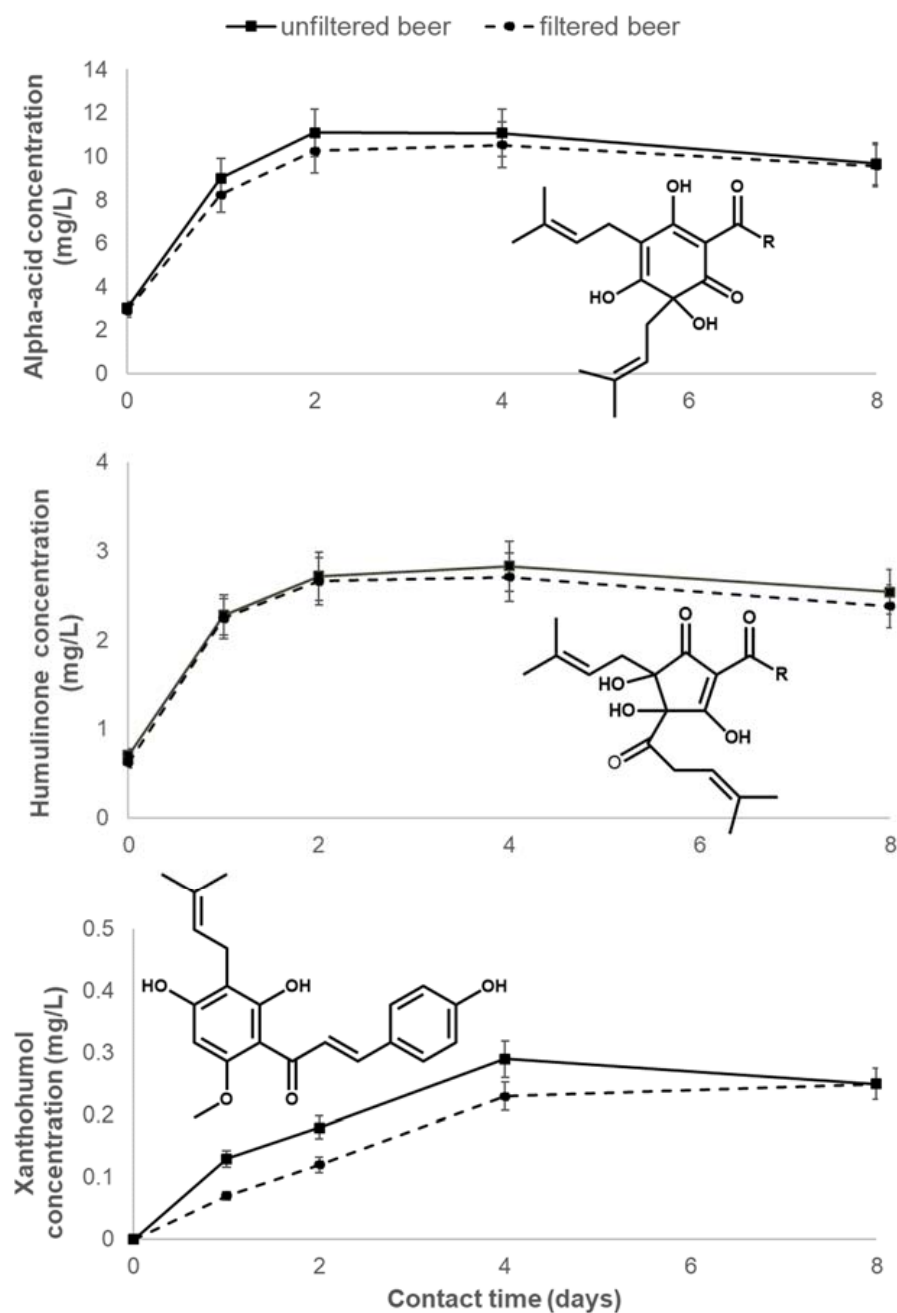


Fig. 4 Hop derived bitter compounds (in mg/L  $\pm$  standard deviation of duplicate analyses) in unfiltered and filtered beers (base beer and dry-hopped beers with different contact time)

panel for evaluation of the beer dry-hopped for 2 days and the beer dry-hopped for 4 days consisted of 10 individuals. All panelists are beer experts for sensory analyses. The beers were evaluated during one session. The scores of all panelists were averaged. Prior to analyses, beers were stored at room temperature.

### 3 Results and discussion

#### 3.1 Beers with Eureka!

With help of Headspace-Trap GC-MS technique, the analysis of different hop derived aroma compounds in filtered and unfiltered beers as well as in base beer, produced in a 20 hL pilot plant,

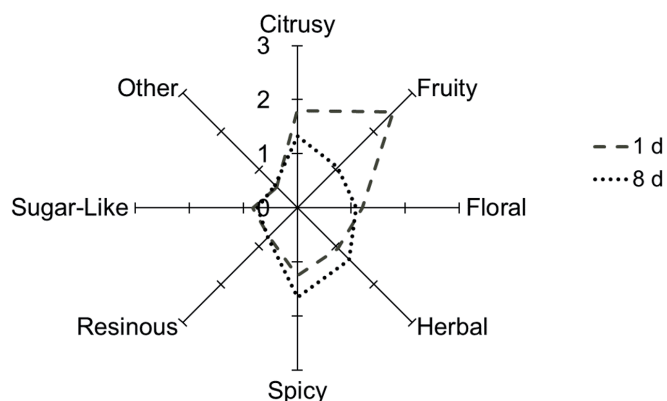
was feasible. The odor-active compounds myrcene, linalool, geraniol, and 2-methylbutyl isobutanoate showed a clear increase from the base beer to all dry-hopped beers.

The highest amount was determined for myrcene with around 1200  $\mu\text{g/L}$  in the unfiltered beers with 2, 4, and 8 days of dry hopping (Fig. 3). The decline after filtration was detected for myrcene only (Fig. 3). The other odor-active compounds were not influenced by filtration. A decrease of myrcene and an unchanged amount of linalool after filtration was also described by *Peifer and Cocuzza* [21]. The amounts of linalool in beers with 2, 4, and 8 days of dry hopping were comparable in a range from 226  $\mu\text{g/L}$  (8 d), 237  $\mu\text{g/L}$  (2 d) to 253  $\mu\text{g/L}$  (4 d). The major transfer was reached after 2 days of dry hopping. These concentrations clearly exceeded the odor threshold value for linalool known from literature (2-80  $\mu\text{g/L}$ ) and the odor threshold of myrcene (9–1000  $\mu\text{g/L}$ ) [6]. Both compounds, linalool and myrcene clearly contribute to the aroma profile of these beers. Geraniol was also detected in the beers which were dry-hopped with the US hop variety Eureka!. In comparison to linalool, the amount of geraniol was lower, in a range between 26  $\mu\text{g/L}$  (1 d) and 38  $\mu\text{g/L}$  (4 d) (Fig. 3). But these concentrations also exceeded the odor threshold of geraniol (4  $\mu\text{g/L}$ ) [2]. The fruity smelling 2-methylbutyl isobutanoate could be quantified with concentrations between 66  $\mu\text{g/L}$  (1 d) and 92  $\mu\text{g/L}$  (4 d). The odor threshold for this compound was described in literature with 78  $\mu\text{g/L}$  in beer [9]. The contribution of this odor-active compound to the aroma profile of tested beers could be demonstrated. For all analyzed hop derived aroma compounds, the major transfer, as already reported for 4-MMP in these beers [5], was reached after 2 days of dry hopping.

To analyze the bitter components, an HPLC-UV system was used. An increase from the base beer was detected for all components tested (Fig. 4). The same behavior as for the aroma compounds could be found for the bitter substances alpha-acids and humulinones. The major transfer was observed after 2 days of dry hopping. The highest amount of alpha-acids with 11 mg/L was determined in beers with 2 and 4 days of dry hopping. The concentrations of humulinones did not exceeded 3 mg/L. The filtration step had no influence on the values of alpha-acids and humulinones. Xanthohumol showed a different behavior. This hop flavonoid increased until day 4 with a maximum amount of 0.3 mg/L (Fig. 4).

In addition to analytical data, sensory evaluations of filtered beers were carried out. Sensory tests were focused on hop aroma as-





**Fig. 5** Sensory evaluation of beer (filtered) with 1 day and 8 days of dry hopping with Eureka! (scale 0-5).

assessment only. The attributes citrusy, fruity, floral, herbal, spicy, resinous, sugar-like, and other were evaluated in all filtered beers. Each individual category includes several sub-divisions for a more detailed specification [20]. In addition to sub-divisions given in [20], black currant-like impression was added to “fruitiness”. All filtered beers were assessed, but to point out the differences in sensory analyses, only beer with 1 day and beer with 8 days of dry hopping are given here (Fig. 5).

Beer with 1 day of dry hopping was characterized as fruity, especially black currant-like, and citrusy whereas the beer with 8 days was assessed more herbal and spicier and less citrusy and fruity. The differences in the analytical data for beers with 1 and 8 days for the two terpene alcohols and the ester 2-methylbutyl isobutanoate are only little but the sensory data showed a clear difference for the attributes citrusy and fruity. The concentration of myrcene in beer with 1 day of dry hopping was significantly lower in comparison to the myrcene amount in beer with 8 days. The higher myrcene concentration in beer with 8 d of dry hopping seems to mask the fruity impression of this beer.

### 3.2 Cold-water extracts with Eureka!

The filtrated aqueous solutions prepared with the hop variety Eureka! were analyzed using HS-Trap GC-MS, GC×GC-TOFMS as well as HPLC-UV. The mean values of triplicates together with the standard deviations of hop aroma and bitter substances are summarized in table 2.

**Table 2** Results of hop aroma and bitter compounds from cold-water extraction (mean value  $\pm$  standard deviation of triplicates)

| Compound                                      | Cold-water extract |
|-----------------------------------------------|--------------------|
| myrcene in $\mu\text{g/L}$                    | $2620 \pm 380$     |
| linalool in $\mu\text{g/L}$                   | $260 \pm 4.2$      |
| 2-methylbutyl isobutanoate in $\mu\text{g/L}$ | $138 \pm 1.8$      |
| geraniol in $\mu\text{g/L}$                   | $36.8 \pm 2.5$     |
| 4-MMP in $\text{ng/kg}$                       | $27.0 \pm 1.7$     |
| alpha-acids in $\text{mg/L}$                  | $18.7 \pm 1.3$     |
| humulinones in $\text{mg/L}$                  | $14.2 \pm 0.50$    |
| xanthohumol in $\text{mg/L}$                  | $0.60 \pm 0.06$    |

For the hop aroma compounds, the highest amount was observed for myrcene with  $2620 \mu\text{g/L}$  followed by linalool and 2-methylbutyl isobutanoate. For linalool, geraniol, and 4-MMP, the cold-water extraction showed a comparable level as detected in the beers from brewing trial with 2, 4 or 8 days of dry hopping (see Fig. 2 and 3). For less polar hop compounds, the actual values in the beer samples were lower. This lab scale prediction tool gives the maximum concentrations of hop compounds derived from a certain hop variety. However, in the actual brewing process these maximum levels are then strongly influenced by various parameters like the type of malt, ethanol content, pH value, contact time and temperature of dry hopping, static or dynamic way of dry hopping, presence of yeast, yeast strain or filtration technology.

## 4 Conclusion

Brewing trials presented that the major transfer of the aroma compounds (myrcene, linalool, geraniol, 2-methylbutyl isobutanoate) as well as of the bitter substances (alpha-acids and humulinones) happened during the first two days of dry hopping with only slight further increase between days 2 and 8. The same findings were reported for the black currant-like smelling hop odorant 4-mercapto-4-methylpentan-2-one (4-MMP) [5]. Sensory analyses showed differences depending on the hop contact time. Increased concentrations of myrcene seems to mask the fruitiness of the beers with longer dry hopping time. Therefore, it is important to keep an eye on the overall hop aroma profile in beer without missing the interactions between different aroma components. With focus on fruity, especially black currant-like, aroma in beer, the received findings suggest a hop contact time of 2 days of dry hopping with the hop variety Eureka!. The longer dry hopping time modified the aroma profile and led to more herbal and spicy characteristics. The presented lab scale cold-water extraction method cannot replace brewing trials which remain essential for the evaluation of dry hopping potential of a selected hop variety.

## 5 References

1. Variety data sheet Eureka!: <https://www.hopsteiner.com/variety-data-sheets/Eureka/>
2. Kishimoto, T.; Wanikawa, A.; Kono, K. and Shibata, K.: Comparison of the Odor-Active Compounds in Unhopped Beer and Beers Hopped with Different Hop Varieties, *J. Agric. Food Chem.*, **54** (2006), no. 23, pp. 8855-8861.
3. Steinhaus, M.; Wilhelm, W. and Schieberle, P.: Comparison of the most odour-active volatiles in different hop varieties by application of a comparative aroma extract dilution analysis, *Eur. Food Res. Technol.*, **226** (2007), no. 1, pp. 45-55.
4. Reglitz, K. and Steinhaus, M.: Quantitation of 4-Methyl-4-sulfanylpentan-2-one (4MSP) in Hops by a Stable Isotope Dilution Assay in Combination with GC×GC-TOFMS: Method Development and Application to Study the Influence of Variety, Provenance, Harvest Year, and Processing on 4MSP Concentrations, *J. Agric. Food Chem.*, **65** (2017), no. 11, pp. 2364-2372.
5. Reglitz, K.; Lemke, N.; Hanke, S. and Steinhaus, M.: On the Behavior of the Important Hop Odorant 4-Mercapto-4-methylpentan-2-one (4MMP) during Dry Hopping and during Storage of Dry Hopped Beer,

- BrewingScience, **71** (2018), no. 11/12, pp. 96-99.
6. Biendl, M.; Engelhard, B.; Forster, A.; Gahr, A.; Lutz, A.; Mitter, W.; Schmidt, R. and Schönberger, C.: Hops – Their Cultivation, Composition and Usage, Fachverlag Hans Carl, 2014.
  7. Fritsch, H.T.: Influence of hops on aroma determining compounds in Pilsner beers and in intermediates of the brewing process (in German), dissertation, TU Munich, 2001.
  8. Fritsch, H.T. and Schieberle, P.: Identification Based on Quantitative Measurements and Aroma Recombination of the Character Impact Odorants in a Bavarian Pilsner-type Beer, *J. Agric. Food Chem.*, **53** (2005), no. 19, pp. 7544-7551.
  9. Takoi, K.; Degueil, M.; Shinkaruk, S.; Thibon, C.; Kurihara, T.; Toyoshima, K.; Ito, K.; Bennetau, B.; Dubourdieu, D. and Tominaga, T.: Specific Flavor Compounds Derived from Nelson Sauvin Hop and Synergy of these Compounds, *BrewingScience – Monatsschrift für Brauwissenschaft*, **62** (2009), no. 7/8, pp. 108-118.
  10. Taniguchi, Y.; Matsukura, Y.; Ozaki, H.; Nishimura, K. and Shindo, K.: Identification and Quantification of the Oxidation Products Derived from  $\alpha$ -Acids and  $\beta$ -Acids During Storage of Hops (*Hulumus Lupulus L.*), *J. Agric. Food Chem.*, **61** (2013), no. 12, pp. 3121-3130.
  11. Maye, J. P.; Smith, R. and Leker, J.: Humulinone Formation in Hops and Hop Pellets and Its Implications for Dry Hopped Beers, *MBAA TQ*, **53** (2016), pp. 23-27.
  12. Algazzali, V. and Shellhammer, T.: Bitterness Intensity of Oxidized Hop Acids: Humulinones and Hulupones, *J. Am. Soc. Brew. Chem.*, **74** (2016), no. 1, pp. 36-43.
  13. Intelmann, D.: Molekulare, psychophysikalische und rezeptorbasierte Studien zum Bittergeschmack von Bier, dissertation, TU Munich, 2011.
  14. Miranda, C. L.; Elias, V. D.; Hay, J. J.; Choi, J.; Reed, R. L. and Stevens, J. F.: Xanthohumol improves dysfunctional glucose and lipid metabolism in diet-induced obese C57BL/6J mice, *Archives of Biochemistry and Biophysics*, **599** (2016), pp. 22-30.
  15. Venturelli, S.; Burkard, M.; Biendl, M.; Lauer, U. M.; Frank, J. and Busch, C.: Prenylated chalcones and flavonoids for the prevention and treatment of cancer, *Nutrition*, **32** (2016), no. 11/12, pp. 1171-1178.
  16. Ferk, F.; Mišik, M.; Nersesyan, A.; Pichler, C.; Jäger, W.; Szekeres, T.; Marculescu, R.; Poulsen, H. E.; Henriksen, T.; Bono, R.; Romanazzi, V.; Al-Serori, H.; Biendl, M.; Wagner, K.-H.; Kundi, M. and Knasmüller, S.: Impact of xanthohumol (a prenylated flavonoid from hops) on DNA stability and other health-related biochemical parameters: Results of human intervention trials, *Mol. Nut. Food Res.*, **60** (2016), no. 4, pp. 773-786.
  17. Van Breemen, R. B.; Yuan, Y.; Banuvar, S.; Shulman, L. P.; Qiu, X.; Ramos Alvarenga, R. F.; Chen, S.-N.; Dietz, B. M.; Bolton, J. L.; Pauli, G. F.; Krause, E.; Viana, M. and Nikolic, D.: Pharmacokinetics of Prenylated Hop Phenols in Women Following Oral Administration of a Standardized Extract of Hops, *Mol. Nut. Food Res.*, **58** (2014), no. 10, pp. 1962-1969.
  18. Analytica EBC 9.47 – Iso- $\alpha$ -acids and reduced iso- $\alpha$ -acids (Rho, Tetra, Hexa) in beer by HPLC – 2010, accessed online, last change 23/10/18.
  19. Analytica EBC 9.49 – Hop aroma components in beer by Headspace-Trap Gas Chromatography (HT-GC) – 2018, accessed online, last change 23/10/18.
  20. Schmidt, C. and Cocuzza, S.: Well-rounded sensory evaluation, *BRAUWELT International*, **32** (2014), no. 2, pp. 116-118.
  21. Peifer, F. and Cocuzza, S.: Influence of filtration on dry-hopped beers (in German), *Brauindustrie*, (2018), pp. 34-37.

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