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Claudia Maurer, Joachim Müller

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Claudia Maurer
Joachim Müller

Institute of Agricultural
Engineering, Universität
Hohenheim, Stuttgart, Germany

Ammonia (NH₃) emissions during drying of untreated and dewatered biogas digestate in a hybrid waste-heat/solar dryer

Digestate from the biogas digestion process contains high amounts of water and nitrogen, which can lead to over-fertilization problems in regions with intensive livestock farming due to a surplus of ammonium, which contributes to the global warming effects. The best option to reduce the water content as well as to concentrate the nitrogen is to dry digestate; however, volatile nitrogen present in the digestate has the tendency to be emitted. Therefore, the objective of this study was to investigate the emissions of untreated and dewatered digestate during the drying process and to determine the nitrogen loss of the drying product by calculating the loss over time by the emissions and compare it with initial and final ammonium content made by chemical analysis. The drying procedure was performed in a hybrid waste-heat/solar dryer. Ammonia rate was measured continuously during drying by Fourier transform infrared spectroscopy. Untreated digestate showed higher cumulative emissions of ammonia (25.9 g_{NH₃} kg_{DM}⁻¹) than dewatered material (7.75 g_{NH₃} kg_{DM}⁻¹) due to lower water content and shorter drying time. Emissions from the filtrate may still be an issue, which should be addressed in future studies.

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1 Introduction

Recently, biogas production developed as a new branch of agricultural activities from animal livestock farming. The German Biogas Association estimated that 6800 biogas plants will be operating in Germany by the end of 2011 with an installed total electrical power of 2559 MW [1]. Since 1992, the number of biogas plants increased 50-fold and this trend is expected to continue further, also including the average size of the plants—which was 370 kW_{el} in 2010 [2, 3]. Most of the biogas plants in Germany are situated in regions with intensive livestock farming [4]. Normally, the nutrient cycle is closed on farm level and there should be no surplus of nutrients. However, most biogas plants additionally digest cosubstrates from neighboring farms or food processing industries to achieve a higher content of organic matter and enhance the biogas yield [2, 5]. Thereby, organic

Correspondence: Claudia Maurer (claudia.maurer@uni-hohenheim.de) Institute of Agricultural Engineering, Universität Hohenheim, Garbenstraße 9, 70599 Stuttgart, Germany

Abbreviations: CHP, combined heat and power unit; DM, dry matter; N_t, total nitrogen content; NH₃, ammonia; NH₄-N, nitrogen in the chemical bond of ammonium

nutrients like nitrogen, as are contained in the digestate, will be accumulated in the nutrient cycle of the farm. In view of the limited quantity of nitrogen that can be applied to the field as regulated by the Fertilization Ordinance [6], it is necessary to find a way to manage and transport the digestate away from the farm and the region, respectively. From an economic perspective, it is not reasonable to transport the digestate for distances longer than 10 km [7]. Due to this limitation, drying of digestate seems to be a viable way to reduce transport costs by mass reduction as well as to discharge nitrogen from the farm's nutrients cycle. Drying digestate has key benefits in handling the surplus of nutrients and the German Renewable Energy Source Act 2009 supports drying of digestate with waste-heat of a biogas plant to produce transportable fertilizer [8]. A negative aspect of drying digestate is the emission of the volatile compounds in the digestate as it contains high amounts of nitrogen in the form of ammonium (NH₄) and ammonia (NH₃), both forms are prone to volatilization. A reduction of ammonium-N content was determined based on results from previous, unpublished studies of the authors. These studies investigated the untreated digestate and the dried digestate with chemical analysis during the drying process in the same dryer.

If the readily available nitrogen in the digestate is emitted as NH₃ during the drying process, the fertilizer value of the

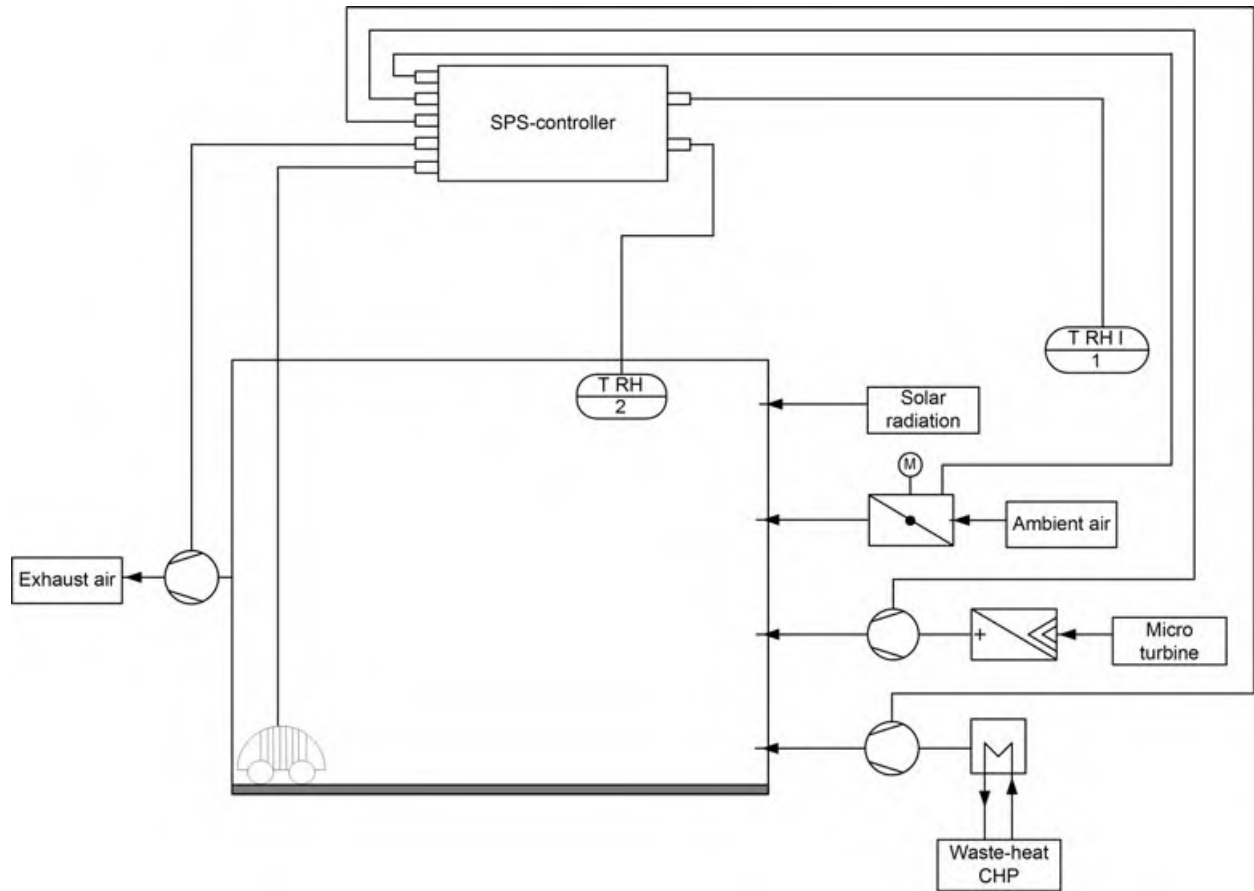


Figure 1. Components and control scheme of the dryer.

product decreases due to the loss of the valuable nutrients. NH_3 in the atmosphere has an indirect effect on climate change via deposition and generation of N_2O . The activity of NH_3 to change the chemical bound leads to deposition of NH_4^+ and contributes also to acidification and eutrophication [9].

Drying of biogas digestate is a frequently discussed issue, however, there is little information about the emissions of digestate during the drying process under large-scale conditions. Hence, the aim of this study was to investigate the emission rate of NH_3 from differently treated biogas digestate during drying in a large-scale greenhouse dryer.

2 Materials and methods

2.1 Hybrid waste-heat/solar dryer

Measurements were performed in a solar greenhouse dryer, using waste-heat of a combined heat and power (CHP) unit (320 kW_{el}) and the hot exhaust gas of a microturbine (200 kW_{el}) of a biogas plant in Kupferzell ($49^\circ 14' \text{ N}$, $9^\circ 41' \text{ E}$) in southern Germany. The drying area of 480 m^2 was canopied with an air bubble film that is transparent for solar radiation and a good insulation to minimize heat losses to ambient. The digestate was spread on the floor and heated by solar energy and energy from CHP unit

and microturbine. Water which evaporated from the surface was transported out of the greenhouse dryer with an exhaust airflow generated by four axial fans installed in the gable. In Fig. 1, a scheme of the dryer is given. The ambient air enters the dryer by the ventilation flap and the exhaust air from the microturbine mixed by ambient air enters the dryer by the inlet fan. The air distribution is supported by the ceiling fans. Heat exchangers in the hall are providing the energy from the waste-heat from the CHP. After taking up the evaporated water, the air is expelled from the drying hall by the exhaust air fan. The ventilation rate is regulated by the programmable controller dependent on the air conditions in the dryer and the drying stage.

The digestate is mixed automatically with a mixing tool up to 12 times per day. The mixing tool is a small robot on four metal wheels with mixing blades installed on two slowly rotating shafts. The robot is equipped with a control system and operates autonomously across the drying area.

2.2 Material

The digestate from a biogas plant operating with wet fermentation technology was used for the experiments. The feedstock of the biogas plant was a mixture of residues from food and feed production, pig, and cattle slurries as well as energy plants.

Investigations were performed with untreated and dewatered digestate, with solid content of 10.5% dry matter (DM) and 22.5% DM, respectively. The DM content was determined by the oven method according to standard method DIN EN 12880 (105°C, 24 h) [10].

The untreated digestate was pumped into the drying hall via a subterranean pipe. The dewatered digestate was filled into the hall with a wheel loader. A mixed sample was collected for the untreated and dewatered digestate as well as for the dried forms of both. Total nitrogen (N_t) was determined according to the Kjeldahl method (DIN ISO 11261) [11] and ammonium nitrogen (NH_4-N) of the tested substrates was determined according to DIN 38406 (photometric method) [12]. Based on these results, the N_t content and the NH_4-N content was calculated for gram per kilogram DM.

2.3 Measurements

The measurements were implemented in accordance with the guidelines from the Association of German Engineers “Realization of stationary source emission measurements” [13]. A scheme of the experimental set up is shown in Fig. 2.

The exhaust air (Fig. 2A) with emissions was channeled through a measurement pipe with a diameter of 920 mm and a length of 7000 mm (Fig. 2D). In this pipe, air samples were sucked through dust filters (Fig. 2E) via a hose to the measurement system. The filters had a diameter of 26 mm and a length of 45 mm. A filling of borosilicate glass microfiber with a penetrability of 2 μm prevented dust particles from entering the measurement system. The dust filters have negligible adsorption effect in terms of nitrogen emissions. NH_3 emissions were measured by Fourier transform infrared spectroscopy (Gaset DX 4000, Ansyco, Karlsruhe, Germany) in an interval of 60 s. Temperature and air pressure were measured with a data logger (EASYLOG 80 CL, Greisinger, Regenstauf, Germany) in the same interval. The airflow rate was calculated by speed frequency

from the characteristic line of the fan, which was calibrated before measurement. Concentration of NH_3 in the exhaust air was calculated to mass concentration:

$$C_{\text{mass};N} = \frac{M_{\text{mol}}}{V_{\text{mol}}} \cdot C \cdot \frac{1}{1000} \quad (1)$$

$C_{\text{mass};N}$ = mass concentration normal (mg/m^3)

C = concentration of emission normal (ppm; mol)

M_{mol} = molar mass of NH_3 (17.03 g/mol)

V_{mol} = molar volume of gas (0.022413 m^3/mol)

Mass concentration $C_{\text{mass};N}$ measured for normal conditions with Gaset DX 4000 was converted into mass concentration for real conditions in the exhaust air for measured temperature and pressure in the measurement pipe:

$$C_{\text{mass};R} = \frac{T_N \cdot p_R}{T_R \cdot p_N} \cdot C_{\text{mass};N} \quad (2)$$

$C_{\text{mass};R}$ = mass concentration for real conditions (mg/m^3)

T_N = normal temperature (273.15 K)

T_R = temperature measured (K)

p_N = normal pressure (1013.25 hPa)

p_R = pressure measured (hPa)

The mass concentration of NH_3 calculated from Eq. 2 multiplied with the measured exhaust airflow Q_t results in the instantaneous mass flow of NH_3 emissions, which was accumulated by adding the values of n measurements during the course of drying and considering the measurement interval of $\Delta t = 60$ s:

$$m_{NH_3} = \sum_n C_{\text{mass};R;n} \cdot Q_n \cdot \Delta t \quad (3)$$

m_{NH_3} = Cumulative NH_3 emission (mg)

Q = Exhaust airflow rate (m^3/h)

Δt = Measurement interval (h)

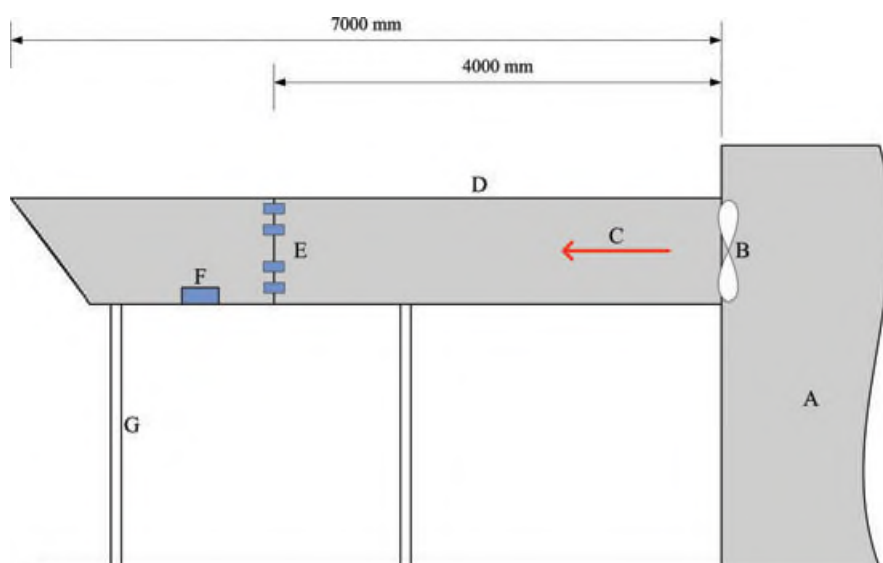


Figure 2. Longitudinal section of the experimental set up; (A) drying hall, (B) exhaust air fan, (C) airflow, (D) measurements tube, (E) filter for emission measurements, (F) sensor for temperature and air pressure, and (G) support.

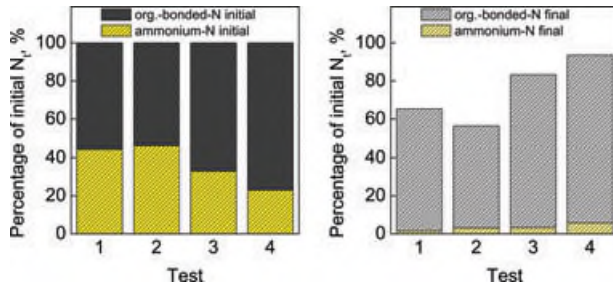


Figure 3. Percentage of total initial nitrogen content (N_t) before (left) and after drying (right) of untreated (tests 1, 2) and dewatered (tests 3, 4) digestate.

3 Results

3.1 Changes of ammonium content during the drying process

The parameters of the drying tests are shown in Table 1. The ammonium-N content of untreated digestate was reduced by dewatering. A separation efficiency of 68% was achieved regarding ammonium-N content, which means that the main part of ammonium-N is captured in the liquid fraction and cannot emit during the drying process. The tests show a significant loss of ammonium calculated on base of the chemical analysis. Figure 3 demonstrates the percentage of ammonium-N relative to the total nitrogen content at the beginning of the drying test (left) as well as the decrease in percentage of total nitrogen in the dried substrates with proportionate ammonium-N content (right). The results show a reduction of the total nitrogen content in the range of 7–43% depending on conditioning of the digestate and required drying time.

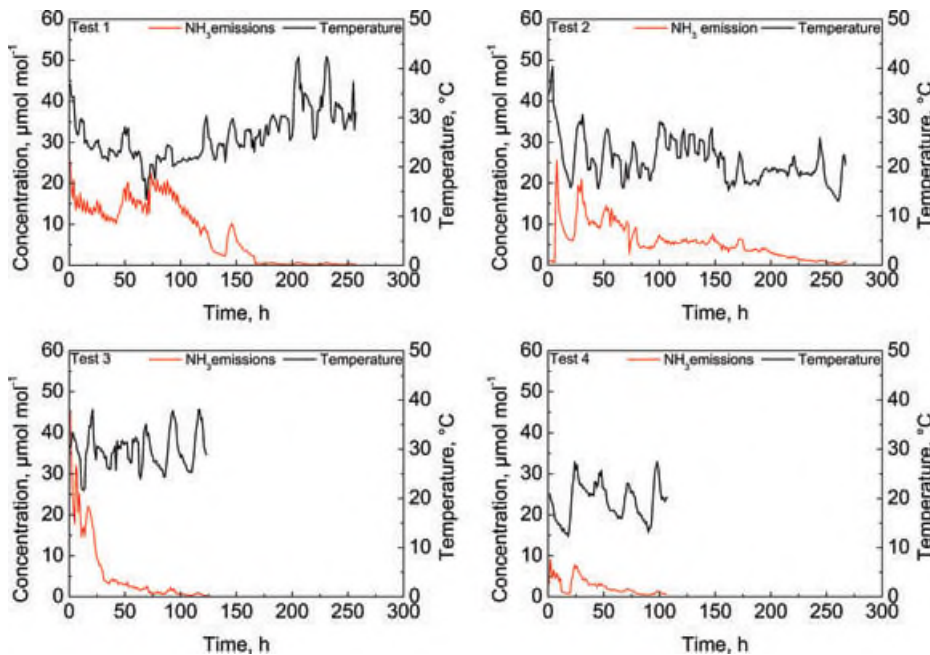


Figure 4. NH_3 emission of untreated (tests 1, 2) and dewatered (tests 3, 4) digestate in the exhaust air of the dryer with the corresponding temperature.

Table 1. Test conditions for drying untreated and dewatered digestate with initial and final mass m_1 , m_2 ; dry matter content DM_1 , DM_2 ; water evaporation Δm_w ; drying time t ; initial and final total nitrogen content N_{t1} , N_{t2} , and initial and final ammonium-N content $\text{NH}_4\text{-}N_1$, $\text{NH}_4\text{-}N_2$.

Parameter	Test 1	Test 2	Test 3	Test 4
Treatment	Untreated	Untreated	Dewatered	Dewatered
m_1 , kg m^{-2}	77.1	93.8	52.1	41.7
m_2 , kg m^{-2}	10.5	14.9	15.4	15.6
DM_1 , %	12.6	9.9	27.1	21.5
DM_2 , %	92.5	62.2	91.9	57.5
Δm_w , kg	31,960	37,831	17,609	12,521
t , h	257	267	123	107
N_{t1} , $\text{gN kg}_{\text{FM}}^{-1}$	5.9	5.5	7.0	5.8
N_{t2} , $\text{gN kg}_{\text{FM}}^{-1}$	28.1	19.6	19.8	14.6
$\text{NH}_4\text{-}N_1$, $\text{gN kg}_{\text{FM}}^{-1}$	2.6	2.5	2.3	1.3
$\text{NH}_4\text{-}N_2$, $\text{gN kg}_{\text{FM}}^{-1}$	0.5	0.6	0.7	0.8

FM, fresh material

3.2 NH_3 emission rate during time

The emissions of NH_3 and the temperature of the exhaust airflow over drying time are shown in Fig. 4. The temperature curve reflects the influence of the solar energy by daily fluctuation between day and night time. During drying, energy input of the waste-heat as well as from the microturbine was almost constant. Drying time for tests with dewatered digestate is shorter due to lower moisture content. The graphs of Fig. 4 indicate the influence of temperature on the emissions of NH_3 . This influence is particularly visible at the beginning of drying. During the first third of drying, increased temperature led to an increase in NH_3 concentration of the exhaust air during drying for both untreated and dewatered digestates.

Figure 5 shows the calculated and accumulated NH_3 emission during drying time. The volatilization of NH_3 with de-

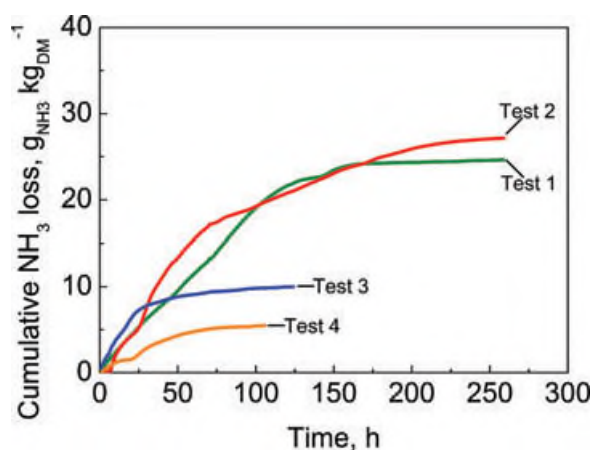


Figure 5. Cumulative NH₃ loss during drying of untreated and dewatered digestate.

watered digestate (7.75 g_{NH₃} kg_{DM}⁻¹) is less than the untreated (25.9 g_{NH₃} kg_{DM}⁻¹). One reason is the low ammonium-N content in the dewatered digestate and another is the higher DM content due to separation.

4 Discussion

Ammonium and ammonia are in a dynamic equilibrium in aqueous solutions. For regulating the emission rate of ammonia, partial pressure differences of the interface between the atmosphere and the surface of the digestate is important as the chemical bond of ammonium changes into NH₃ and can volatilize into ambient air [14, 15]. The individual tests demonstrated the volatilization of ammonia during the drying period. Deng et al. showed that the emission rate is decreasing with elapsed drying time, so the decreased concentration of NH₃ was expected [16] as shown in Fig. 4. The influence of temperature to the emissions of ammonia is higher at the beginning of the drying process due to the higher amount of volatile chemical nitrogen bounds in the digestate. The temperature in the measurement tube reflected the temperature in the dryer and in other studies temperature is described as one factor influencing the emission of ammonia [17, 18].

In order to avoid volatilization of NH₃, different options have been successfully realized in the last years like chemical, microbiological, and mechanical ones, but these studies investigated the NH₃ volatilization in manure or wastewaters [18–22]. Dewatering the digestate before drying enhances the solid content and the total emission potential of NH₃ is reduced as shown in Fig. 5. In this study, an ammonium-N separation rate of 68% was achieved by dewatering, i.e. 68% of ammonium-N was captured in the liquid fraction. Thus, dewatering digestate prior drying process is a way to minimize the NH₃ emissions during the drying process. However, the separated liquid fraction has still high water content and is not appropriate to transport for longer distances. In terms of total nitrogen content there is a concentration of total nitrogen in the dried digestate regarding the output after the drying process. The dried digestate has in av-

erage 28.85 g_N kg_{FM}⁻¹ in untreated digestate and 17.2 g_N kg_{FM}⁻¹ in dewatered digestate. Ammonium-N losses up to 97.5% for the untreated digestate and 91.7% for the dewatered digestate were measured.

Further support of the use of waste-heat from CHP to dry digestate for use as fertilizer will be provided for the Amendment of Renewable Energy Source Act 2012. Consequently, an increasing number of new installed drying plants is expected. Further research is necessary to investigate the influence of the drying temperature and water evaporation on the NH₃ emission rate of untreated and dewatered digestate.

In general, drying of digestate seems to be a viable way to handle the digestate if it is necessary to enhance the transport efficiency or to reduce storage requirement. Nevertheless, it should be ensured that the ammonia can be recovered or the emissions can be reduced to a minimum.

Practical Application

Handling the biogas digestate is an essential issue in the management of biogas production systems. The German Renewable Energy Source Act 2009 supports drying of digestate by using the waste-heat of CHP for producing organic fertilizer. The fertilizer value of the digestate depends on the nitrogen content and therefore losses of NH₃ by volatilization should be prevented. NH₃ emissions to the atmosphere have an indirect effect on climate change via deposition and generation of N₂O. The deposition of nitrogen is also contributing to acidification and eutrophication. This study investigates the effect of dewatering prior to drying and gives a comparison of the emission rate during drying of untreated and dewatered digestate. Results showed that most of the ammonium volatilized during the drying process no matter if the digestate was dewatered or untreated. Though, dewatering the digestate emissions of NH₃ are lower, fertilizer value of the dried digestate is higher and global warming potential is reduced.

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