Once more the lure of 'unexplored' country and prospects for pastoral and agricultural expansion were the impetus for Donald Mackay’s Arnhem Land expedition in 1928. Again, Herbert Basedow and Frank Feast joined Mackay, along with Walter Sully, a cinematographer, and CW Lovell, who was responsible for stores and equipment. They departed Sydney on 3 April on the SS *Marella*, stopping at Brisbane where they purchased a buggy, and arrived in Darwin on 14 April. Two days later the party caught the train south to Katherine, where they took delivery of 26 horses and 6 mules, and Larry Coonan was engaged to look after them. Two Aboriginal men, Barney and Deacon, were also hired to assist.

Basedow found the horses and mules initially problematic, noting 'some exciting scenes when saddling and loading them. Many outlaws are included among our “mob” which was guaranteed quiet and well broken in’. By mid-afternoon on 18 April everything was ready and the party headed east for Roper Bar. The animals continued to play up but eventually settled. At Roper Valley station, owner John Rogers provided 'much information' on the country in Arnhem Land. It was probably Rogers who advised them against taking the buggy any further than Roper Bar. As Lovell had been hired to drive the buggy, his services were no longer required and he departed.

On 5 May the party crossed the Roper River and headed north to the Wilton River, which it followed for several days. After passing Mount Marumba, the expedition made for the Goyder River, passing through low hills and eucalypt forests with occasional paperbark thickets. Some time after crossing the Goyder a hill was climbed to examine the lie of the land. They decided to turn north-west. Prior to Mackay’s expedition, only a few exploring parties had travelled through the region, so a comprehensive map was not available. Typically, the way forward was determined by climbing a vantage point to assess the likely route ahead. After rejoining the Goyder, the party reached Mount Delight on 20 May and then headed north-west again. Two days later they struck the Blyth River, following it upstream before making a particularly difficult crossing.
On the afternoon of 25 May, they came upon a small family of Aboriginal people, the first they had seen since leaving Roper Bar. As they continued on their westerly path, the terrain became increasingly rough. After crossing the Cadell River they encountered a maze of rocky gorges. Turning north, they struck the Liverpool River near its headwaters, surprising a large camp of Aboriginal people. Basedow documented what he could in the short time they remained there:

We make for one group consisting of old man, lubra and piccaninnies, but as we approach all except the old man decamp as do all other groups ... An old blind man left in camp ... keeps the flies from his face with the 2 wings of wild goose. At length a warrior returns to camp who has been out hunting. His body covered with earthy ochre all over, large bundle of spears, a long rock python. He is friendly and soon another hunter with his lubra and piccaninny return ... They manage to coax all the fugitive members back, including the old man who it appears is the chief. When we disturbed him this morning he was making very finely plaited dilly bag with strands dangling from its side upon which feathers of Blue Mountain parrot tied. This appears to be a distinction mark of rank. Two of the men express themselves as willing to accompany us to Oenpelli and show us the best way over the rocky ranges.

The two men who undertook to guide the party suddenly departed after a few days of travel and once again the expeditioners were on their own in difficult country. Before they reached Oenpelli, several horses became bogged while crossing a stream and much of their dwindling food supply was spoilt. On the way, Basedow photographed and sketched rock paintings. At one site, just east of Oenpelli, Basedow recorded a painting of a figure that he attributed to 'the deity of lightening, rain and thunder known as Mérenn (Kakatu) or Namarruwan (Jingu)'.

On 9 June they met another Aboriginal man who guided them to Oenpelli, where they arrived later that day. Here Basedow visited Aboriginal camps, where he noted a man with a 'broken scalp and smashed finger, result of tribal brawl over a woman' and made other medical inspections. The next day they departed for Pine Creek, crossing the South Alligator River before arriving at Goodparla station. From Pine Creek they caught a train to Darwin and sailed for Adelaide on 8 July, again on the Marella.

Basedow’s and Mackay’s assessment of the economic possibilities of the country they explored was pessimistic. Grassed land was either too heavily timbered or small and scattered, and forests suitable for timber were too small and isolated to permit harvesting. Only one potential area for goldmining was identified.

Sully produced a travelogue from his film footage, titled 'Mysterious Arnhem Land', which was shown around the world in 1928. Unfortunately, neither the film nor other footage from the expedition has been located. However two sets of photographs, by Basedow and Feast, as well as a handful of Mackay’s, remain to provide a visual record of the expedition.